AN ASSESSMENT OF EFFECTIVE THERMAL CONDUCTIVITY OF EPOXY-TiO_2 COMPOSITES

A THESIS SUBMITTED IN PARTIAL FULFILLMENT OF THE REQUIREMENTS FOR THE DEGREE OF

Master of Technology

in

Mechanical Engineering

(Thermal Engineering)

By

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NATIONAL INSTITUTE OF TECHNOLOGY ROURKELA

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CERTIFICATE

This is to certify that the thesis entitled, ***"An Assessment of effective thermal conductivity of Epoxy-TiO_2 composites"*** submitted by **Mr. Sumit Bhanariya (213ME3426)** has been carried out in partial fulfillment of the requirements for the award of Master of Technology Degree in **Mechanical Engineering** with specialization in **Thermal Engineering** during the session 2014-2015 at the National Institute of Technology Rourkela is an authentic work carried out by him under my supervision and guidance.

To the best of my knowledge, the matter embodied in the thesis has not been submitted to any other University/Institute for the award of any degree of diploma.

Place: NIT Rourkela

Date:

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ABSTRACT

The development of microelectronic devices like PCB,electronic packaging etc. requires high thermal conductivity materials for fabrication. This provoke the needs of development of particulate filled polymer matrix composites (PMCs) that possess high thermal conductivity. This research provides a potential material system to address this problem. An analytical model is proposed to estimate the effective thermal conductivity of polymer composites filled with cubical particulate fillers. By using the mathematical correlation developed by this model, effective thermal conductivity is calculated and compared with values obtained from models by previous authors in this regard for different volume fractions (13.4 and 26.8 vol.%) of cubical shaped TiO_2 particulates. Unitherm^{TM} 2022 tester is used to measure the effective thermal conductivity (k_{eff}) of epoxy-TiO_2 composite for these volume fraction as per ASTM E-1530. These values of effective thermal conductivity (k_{eff}) obtained from the experiment are compared with these model developed in previous literature and the proposed model. It is found that the the values of effective thermal conductivity calculated by proposed model are very near to the experimental values of effective thermal conductivity.

Keywords-PMCs,particulates,effective thermal conductivity,volume fraction.

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Chapter 1

INTRODUCTION

1.1 Permeable

Power density of the electronic device is quite high which causes the generation of heat within the electronic component. Thus it is required to carry away the heat from the electronic components and prevent the device by maintaining the temperature lower than the critical values [1]. Traditionally heat sink which were metal based and having high cost were used to dissipate the heat generated within the component which are not acceptable nowadays because of thermal cracking[2]. Also the weight of these devices is also the main consideration. Low weight electronic devices are the main requirement in today's world so as to carry them from one place to other place. These requirement led to increase in the demand of high performance micro-electronic devices enforcing for the requirement of the advancement in the technologies regarding electronic packaging. The wide application of electronic and electrical technologies and their device requires the better micro-electronic packaging. Micro-electronic packaging is the multi-discipline subject in the field of electronic engineering. It considers so many issues like cost, mechanical properties, heat transfer characteristics, reliability etc. [3]

Polymers and ceramics are generally in demand for packaging materials because of their better mechanical, electrical and thermal properties [4]. Generally available polymer for packaging material are polypropylene (PP), polyethylene (PE), polyamide (PA), epoxy, polyimide, However, common polymers for packaging, such as polyester, polyethylene (PE), acrylonitrile-butadiene-styrene (ABS)etc. They have less thermal conductivities and high coefficient of thermal expansion (CTE) thus they cannot provide effective heat flow which led to thermal failure.

It is always demanded in the world of advancement to replace existing materials by advanced material having better properties to achieve the new requirements. The use of larger particle and surface treated filler resulted in composite materials with enhanced thermal conductivity. Carbon based filler material also used as filler material [5, 6], but they couldn't give satisfactory results in the case of thermal conductivity. Ceramic filler materials like SiC, Al_2O_3, Si_3N_4 etc. also increases the thermal conductivity.

Incorporation of metallic powder also increases the thermal conductivity of composite [7-9] but it is found that weight of the composite material may also increases more than required. Also with the use of metallic powder electrical conductivity also increases which is not required sometimes. Traditionally used metal based metal alloys like copper, aluminium and all other metal alloy are not able to possess ability to be used in electronic packaging [10], thus polymer composite material came into existence to take care of such issues. For fabrication of such particulate filled polymer composite the characteristics like low material density, low coefficient of thermal expansion, low electrical conductivity, higher thermal conductivity etc. are taken into consideration [11].

1.2 Introduction to Composite material

1.2.1 Composite Materials

Composite are formed by the combination or mixture of two or more different materials. In composite material there is one reinforcing phase, present in the form of either particulates or fibre or both and the other one is matrix phase. This reinforcing phase is embedded in the matrix phase.

This combination of reinforcing phase and matrix phase gives the predominant properties which are generally not exhibited by a single material. The matrix phase is provided to transfer the forces among the reinforcing particles or fibre and to give them protection from atmospheric and mechanical damages whereas. Thus the composite provides the s synergistic effect of both the phase.

The composites is not human invented term. There are so many composite which are naturally found in nature. Wood is the better example of natural composite material in which polymer resinous matrix, polysaccharide lignin is composed of cellulose fibres which provide wood the good mechanical properties and thermal insulation. Thus the fabrication of composite material does in such a way that to maximize their good properties which are needed and minimize their bad effect so that their performance may increase. This optimization should also be base d on the cheaper composition and fabrication, simpler method and better results.

A structural of composite material gives the reasonable effect on the properties of composite material. Also the distribution of material and particulates characteristics also affect the properties of whole composite material. If the reinforcement of material is non-uniform then the particles are heterogeneously distributed which increases the possibility of failure of weak areas. If the orientation of particulate changes than the anisotropy of the system also changes.

The reinforcement of short fibres and particles gives low and moderate performance because the reinforcing fibre/particle gives local strength and the matrix phase takes the whole load of the composite whereas the reinforcement of continuous fibre and particles provides high performance to composite material. The continuous distribution of particles and particles provides them strength and matrix phase gives safety to the particles and fibre and transfer load among them.

1.2.2 Classification of composite materials

On the basis of form of constituents, two types of composites are classified: composites with particle and composites materials with fibre

a) Composites with Particle

When a composite material is reinforced with particle then it is known as particle composite. Particles are used to increase some properties of material like stiffness, resistance to abrasion, temperature, decrease of shrinkage etc. In some of the composites particles are also used as a filler material to reduce the effective cost of material without compromising with its characteristics. The particle-matrix combination are chosen on the basis of properties required. For example, brittle metal incorporated in ductile metal to improve their properties at elevated temperature while preserving ductility.

b) Composites with fibre

When a composite material is reinforced with fibre then it is known as fibre composite. The primary function of fibre in the composite is to increase the strength of the composite material. Its individual property corporates in the increase in the property of composite material.

On the basis of their extraction

a) **Natural composites -** Composites which are produced by the nature or present in the nature are called natural composites.

Example are concrete, wood, asphalt etc.

b) **Synthetic composites -** These are the composites manufactured in the industry in which one material is combined to other material in a controlled manner to get the desired properties, structure and geometry.

Synthetic composite are generally used to achieve the desired mechanical properties required for a particular application.

Based on this the composites are classified in two parts:

- **Micro-composites**
- **Macro-composites**

Micro-composites - In micro-composites the size of the component in the composite are very small like atomic size. Metallic alloy having multiphase structure are the best example of the micro-composite. In micro-composites the component having the atomic size are finely distributed in the material.

In the macro-composites the size of the component is large compare to micro-composites.

Macro-composites are divided into two parts-

a) Large particle composite
b) Fibre-reinforced composite

In large particle macro-composite, large size particles of single type of material is uniformly distributed in the matrix. The matrix generally used are polymer, metal or ceramic. Cermets in which ceramic particle incorporated in metal matrix is the example of the large particle composite. Cermet are use as cutting tools in machining. For better and effective strengthening, the size of the particle should be small and distributed uniformly throughout the matrix.

Fibre-reinforced composites are prepared to increase the strength and modulus of elasticity without enhancing the specific gravity. Low weight is also the main characteristics of the fibre-reinforced composites. In Fibre-reinforced composites, the load is transmitted and distributed to the fibre through the matrix. For effectual transfer of the load, the bond between the matrix and fibre must be very strong.

Fibre-reinforced composites are classified into two major parts-

I. Continuous fibre composites
II. Short fibre composites

Continuous fibre composites-

Continuous fibre composites are the composite fibre in which the fibre which is to be reinforce are long and extended over the full length of the material. The mechanical properties of such composites are different in different direction i.e. they are anisotropic. In the direction parallel to the fibres, the tensile and compressive strength of Continuous fibre composites is very high whereas in the direction perpendicular to the fibre, it is vice versa.

Short fibre composites-

In short fibres composites the length of the fibre is too short so it doesn't help in the increase in the strength of the composite material. But the mechanical properties of such composite doesn't vary with the direction. Because of the isotropic nature of the short fibres composites, they are useful in the application in which stresses are applied along so many direction.

On the basis of matrix material

1. Polymer-matrix composite
2. Metal-matrix composite
3. Ceramic-matrix composite
4. Carbon-carbon matrix

(i) Polymer matrix composite

Since Polymer doesn't possess adequate mechanical properties for so many structural Purpose. They also have low stiffness and strength. Thus these properties are fulfilled by incorporating filler material into the polymer matrix. Formation of such polymer matrix composite material are quite simpler, it doesn't require high pressure or high temperature for its processing. Thus polymer composite are quite cheaply and rapidly developed. Commonly used matrix material in polymer-matric composite are polyester and vinyl ester. Glass, carbon and aramid are used in such matrix as the fibre material and boron (BN), aluminium oxide (Al_2O_3), and silicon carbide (SiC) are used in the application where strength is needed. Polymer matrix composite also have greater elastic modulus that the base polymer and they are not brittle as ceramic. Application like aircraft and space craft uses polymer matrix composite as material.

Polymer matrix composite are further classified on the basis of reinforcing material as fibre reinforcing polymer (FRP) and particle reinforced polymer (PRP).

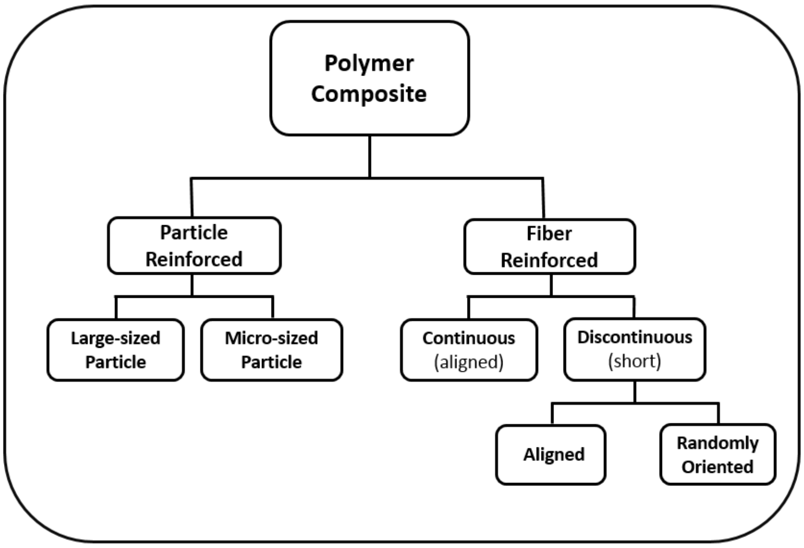

Figure 1.1 Classification of Polymer composites based on reinforcement

(ii) Metal matrix composite

In metal-matrix composite metal is used as matrix material. Metal matrix composite possess better properties at elevated temperature, low coefficient of thermal expansion, higher specific strength, higher thermal conductivity, higher creep resistance, dimensional stability, and higher specific modulus than the base metal material. They also have higher operating range and higher specific strength than the polymer matrix composite material. They are more costly than PMC's.

Generally used matrix material are alloys of different metal like aluminium, titanium, magnesium, copper, zinc etc. Nickel and Cobalt based alloys are used as matrices for high strength and high temperature operating range. Carbon and alumina are used as fibre and alumina and silicon carbide particles used as filler material.

They are widely used in so many bigger applications like structure of submarine and aerospace, space shuttles and IC and SI engine components.

(iii) Ceramic-matrix composite

Ceramic-matrix composites are designed to possess higher properties and elevated temperature, higher creep resistance and to increase the toughness of the material. But they also have tendency to brittle fracture. Generally used matrix material in Ceramic-matrix composite are alumina and zirconium oxide. And zirconia, alumina and silicon carbide whisker are used as filler material.

(iv) Carbon-carbon composite

In Carbon-carbon composite, carbon is used as both matrix material and reinforcing agent. The processing method of such composite are quite expensive and complex than the other composite material. They have high mechanical properties and find application in automobile parts, turbine and engine parts, space shuttle etc.

1.2.3 Applications of Composite Material

Composite materials are most extensively used materials due to its advantageous over other material like it has availability, adopting nature in various situations and it can easily combine with other material to form different material with desirable properties to serve particular purposes.

Automobile Engineering

The most important advantage of composite material to use it as light weight material. It has a higher **strength weight ratio** than other material. Light weight, easily availability of materials, changes in trends, shock absorbing capacity, strength, material selection are the basic requirements of the Automobile industry. Number of components of car, buses, trucks etc. like doors, window, chassis, and so many mouldings uses composite material. Composite material like Glass reinforced plastics, polyester resin are also cost effective. Glass and Sisal fibre are used in so many automobile parts. In road transportation polyester resin are used as a filler material. Polyester can easily be designed, can produces so many parts. It also has low cost and easy availability. In heavy transportation vehicles like trucks, dumpers resilience and reproductivity are required which can be provided by composite material only. When Thermoplastics reinforced with fibre can also be used in vehicle parts because of its higher mechanical strength. Thermosetting resins are used in paint finishing of so many vehicles parts. Cellular cellulose acetate foam cores covered by fibre glass are used in the accessories of cars to provide them strength and rigidity.

Aeronautical and Aerospace Engineering

So many parameters are considered during the production aeronautical and aerospace parts like speed, weight, safety and power. These parameters requires selection of material that provide the optimum values of these parameters. This lead to prediction of composite material which could be suitable for these parameters. Design parameters are also required for such structure. But initially importance is given to production parameter. Aeroplane parts like air frame, spoilers, tail-plane structures, wings, rotor blades, fuel tanks, drop tanks, propellers, doors, seat components, windows, air distribution ducts uses glass fibre composites, resin-impregnated honeycomb core material for their production. Carbon-fibre reinforced material used in jet engine, rocket nozzles, disc brake because of higher temperature in these parts. Performance of aircraft engine can also be increased by the use of Fibre epoxy composites.

Electronics and Electrical Engineering

Advancement in the electronics and electrical industry requires the composite material adoptable in number of issues. Structure of electronics device requires high strength, high thermal conductivity, low coefficient of thermal expansion, high modulus, low or high electrical conductivity, low dielectric constant of the material. Electrical component like generator, switch gear requires high strength insulation, thermally stable, moisture resistant material. These issues led to the development of composite material. Hybrid-Polymer composite material are used for such factors. Substrate material like ceramic, AIN, Al_2O_3 are used as filler material chips for interconnection.

Biomedical Engineering

Various factors like strength, Poisson ratio, hardness, flexural modulus, elastic modulus are considered during the selection of material in biomedical application [053]. Carbon fibre epoxy, carbon fibre/ultrahigh molecular weight polyethylene, silica/silicon rubber etc. are the examples of polymer composite biomaterials. Carbon-fibre reinforced plastics and carbon containing materials are used in artificial body parts purposes such as in fracture fixation plate, arthroplasty in knee (hip replacement), in mandible, joint replacements, external support etc. Heart valve component are made by pyrolytic carbon based composites. Polyethylene used in synthetic bone graft. Thermoset polymer composite reinforced with glass, carbon or Kevlar fibres are used in prosthetic limb.

Marine Engineering

Fibre reinforced polymer composite material used in the boats and boat builder, Glass reinforced polymer composites are used in marine craft because of its easily availability, excellent corrosion resistance and light weight which results in decrease in the fuel consumption. Transport pipelines, fishing boats, transport vessels, cooling circuit uses GRP fir its construction. For safety purpose also so many polymer composite are used in off shore structure. Composite materials are also used in fire protection piping circuits, floorings ladders, tanks, accommodation parts, blast panels etc. High compression properties application submarine, oil exploration, military tanks also uses composite material.

Sports goods

Sports goods industry also accept the use of composite material. So many industries are trying to take the advantages of new material like boron and carbon fibre composite despite the traditional woods and metal in sport industry. So many sport equipment like tennis rackets, golf clubs, fishing rods, cricket bats, hockey sticks, archery, baseball bats use composite material for their production.

Chemical Engineering

So many chemical industries using polymer composite reinforced with both filler and fibre in chemical plant equipment like pumps, valves, containers, piping, pressure vessels etc. These composite are generally moulded.

Domestic appliances

Increase in the cost and weight of metal and its alloy leads to development of composites material for the use I domestic appliances. Polyester moulded compound and thermoplastic are the most common composite material used in the domestic appliances. In all kind of kitchen equipment, Almira, refrigerator, television, helmets, furniture, different types of casing and so many appliances, composite materials are used.

Thus composite materials has found experienced a rapid growth in last decades because of their (a) easy fabrication (b) light weight (c) higher strength to weight ratio, (d) corrosion resistance, (e) higher impact resistance, (f) flexibility in the design, (g) stability in the dimension, (h) durable. Future prospects for composite material are quite acceptable due to all the advantageous. Also the automation is going to be introduced in the field of composite material thus the fabrication is becoming less expansive and simple and the market is expanding. So many work has done on fibre reinforced polymer but less attention is drawn on the particulate filled polymer composite material. Thus this present work is undertaken for particulate filled polymer composite in order to study its properties and their effect.

1.3 Introduction to the Topic

The present work focusses on the thermal behaviour of the polymer composites filled with particulate. Fabrication of such composite is also done to conduct experimental test on them for the further investigation. Continuous arise in the power density of electronic packaging results in the need for superior material having high thermal conductivity. They should have enough thermal conductivity to carry away the heat generated by electronic device and low coefficient of thermal expansion. Furthermore, low weight of such material is also desirable in some of the application. In electronic devices an network of small component and it is integrated inside the packages like IC (integrated circuit), semiconductor, Hybrid integrated circuit (HIC) etc. Thus the packaging material should approach to such design.

Conventional material used so far couldn't meet with the all these requirements of electronic packaging material. Thus the multifunctional material are need to be investigate which possess all these properties which are suitable in such applications.

Neat polymers like epoxy or polyester were commonly used to electronic application but they have low thermal conductivity and high coefficient of thermal expansion which causes thermal failure like cracking etc. Reinforcement of metal powder like aluminium and copper in these polymer led to increase in the thermal conductivity. But with the reinforcement of such materials, the electrical conductivity may increase, thus possibility in the increase in the dielectric constant may rise which affects saviourely due to which ceramic are proffered over all other material like metal powder to use as particulate in polymer matrix.

For characterization of thermal behaviour of these composite, thermal conductivities considered to be most important parameter. Thus it becomes important to calculate the effective thermal conductivity. In this work, a theoretical model and its corresponding mathematical correlation has been for estimation of effective thermal conductivity these composites.

For last 30 years, epoxy had played an important role in the electronic application. They were used in the hybrid industry in sealing packages, protecting devices and as components bonding. Because of their attractive nature in the alternative to the eutectic bonding in hybrid, they are used in the packaging in the semiconductor. Nowadays, solvent-less conductive epoxies are used in plastic encapsulated devices in 80 % of die attach market of the whole world. The reason behind the spreading in the use in the epoxy in electronic packaging are: their profile are generally cured at low temperature which are suitable for the component which are temperature sensitive, gives excellent performance at higher temperature, possess better mechanical, thermal and electrical properties, doesn't contract while cooling, having excellent resistance to wide chemicals, having adhesive property to immense variation of fibres, which are desirable in these applications.

Micro-sized titanium dioxide (TiO_2) is taken as the filler material in the epoxy in the present investigation. TiO_2 with a moderate thermal conductivity and low CTE is a potential filler material to be used in polymeric matrices. The objective of this work is evaluate the effective thermal conductivities of these composites by analytical as well as experimental methods. Through addition of highly conductive inorganic filler into the epoxy matrix, it is possible to achieve a better heat conduction path and decrease the thermal contact resistance at the filler–matrix interface that attributes to increase in thermal conductivity.

Chapter 2

Literature survey and Theoretical Background

2.1 Literature survey

In this chapter, the summary of different literature observed within the period of this work are described. In this chapter the background information related to present work is provided. In this chapter, the review are provided on the following topics:-

- Polymer matrix composites filled with particulate
- Thermal conductivity of composites filled with particulate
- Models for calculating Thermal conductivity of composite materials
- Thermal conductivity of Hybrid filled polymer composites
- Composites material filled with TiO_2

2.1 Study of Polymer matrix composites filled with particulate

The future materials are expected to possess high heat dissipation capacity, high conductivity, low coefficient of thermal expansion etc. Thus so many advanced materials have been drawn for encountering such problem. These materials may be metal matrix composites, ceramic matrix composites, carbon-carbon composites, polymer matrix composites (PMCs) [12] Polymer matrix composites are using extensively in many industries because of their characteristics being fitting in different application. These characterizes high thermal conductivity, high corrosion resistance, increase in mechanical properties like wear resistance and hardness [13] ease of process-ability, durability at elevated temperature[14], are high corrosion resistance, low density, low cost[15-17] etc. Different types of filler material are included in polymer matrix in order to get the desirable properties. Particles generally used for reinforcing are magnetic powder [18], mineral powder, ceramic particles, metal powder like aluminium, copper, zinc, amorphous material etc. Some of the particulates used to increase the thermal conductivity of the composite material required in the various application like in electronic packaging but sometimes it is also required to decrease the thermal conductivity to use the material as an insulating material.

Bonner [19] found that when the polymer matrix included with the micro-sized particulate, the positive effect are seen when the filler content i.e. volume fraction is greater than 20% but it is also seen that above this volume fraction some of the important properties of polymer matrix composites like aging performance, density, processability also affected.

In so many literature [20, 21], it is found that with the inclusion of silica particulate into polymer matrix leads to increase in mechanical, electrical and thermal properties. The effect of variation of size, shape, specific surface area and volume fraction also seen on the variation of mechanical properties of composite materials by many researchers. Adachi et al. and Moloney at al. [22-25] described the variation of mechanical properties with the variation of volume fraction of particles.

Zunlong et al. [26] studied the effective thermal conductivity of graphite particles and carbon fibre filled composite material and seen the impact of the volume fraction, size and distribution style of the graphite particles on the PTFE composite material and optimum volume fraction is obtained.

Agrawal et al. [27] studied the thermal conductivity of Polymer composite reinforced with micro-sized aluminium nitride particle with volume fraction of 25%. It is seen that Incorporation of these micro-sized aluminium nitride (AIN) increases the effective thermal conductivity by 446%.

Leung et al. [28] researched on the development of polymer composites. He developed the composite material filled with spherical particulates and developed a theoretical model for estimation of the effective thermal conductivity of the composite material. They also designed a thermal conductivity analyser to calculate the effective thermal conductivity of composite material experimentally.

Inorganic mineral filler has a reasonable effect on polymer. Inclusion of them in polymer matrix leads to increase in the stiffness and reduction in cost. To avoid detrimental effect and get the simultaneous feature of these co0mposite is also the main cause to use such filler material. Characterization of inorganic filler material filled in polymer matrix composite is also done by determining the particle size and shape [29, 30].

Silica nanoparticles are filled in polymer matrix and the effect of its volume fraction and particle size is seen in the on the toughness of polymer matrix composite. Silica nanoparticles having the different diameters 23, 74 and 170 nm are used in Pepperdine-cured epoxy polymer. It is seen that by varying the particle size there is no effect on the toughness of the composite but it increases with increase in the concentration of the particle [31].

2.2 Study of thermal conductivity of composites filled with particulate

Most of the studies is done regarding the heat transfer in polymer by Hansen and Ho [32], Peg and Landel [33], Henning and knappe [34]. Some of the work is done on heat transfer in polymer is done to increase the thermal conductivity by varying the molecular orientation but it is not always favorable to increase the thermal conductivity by varying the orientation. Thus addition of thermally conductive filler material and fibre in polymer is the simplest and practical means to improve the thermal conductivity.

Some of the studies are done to study the effect of fillers shape and size and orientation on thermal conductivity of the composite material [35].

Banjare et al. [36] described the physical and thermal characterization of composite material reinforced with red mud particulates. They studied the effect of filler particle's volume fraction on the effective thermal conductivity of these composite materials. By the addition of 25vol% of red mud filler, there is 135% increase in the thermal conductivity of these composite material.

2.4 Models for calculating Thermal conductivity of composite materials

So many theoretical models and correlation have been developed by number of researchers to calculate the effective thermal conductivity of composite material. Also models are developed for the two phase mixture.

In Maxwell model 1873 [37], the distribution of molecules is considered to be randomly distributed and having spherical shape which are non-interacting homogeneous and dispersed in homogeneous medium. The expression is shown below.

$$k_c = k_m \left[\frac{k_p + 2k_m + 2\phi(k_p - k_m)}{k_p + 2k_m - \phi(k_p - k_m)} \right] \tag{2.1}$$

So many articles [38, 39] describes the models for the estimation of effective thermal conductivity of a two-component composite. In the basic model the effective thermal conductivity is calculated by considering the material in which the molecules are arranged either in series or parallel with respect to the direction of heat flow. For these arrangements fundamental correlation has been given.

Series heat conduction model:

$$\frac{1}{k_c} = \frac{1-\phi}{k_m} + \frac{1}{k_p}$$

Parallel heat conduction model:

$$k_c = (1-\phi)k_m + \phi k_p$$

Where k_m, k_p are the thermal conductivities of the matrix and the particulate respectively, k_c is the effective thermal conductivity of the composite and ϕ is the volume fraction of the particulate.

Lewis and Nielsen [40] proposed a semi-theoretical model by using the Halpin-Tsai equation of two phase mixture in which they also considered the shape and orientation of particle. The particulate reinforcement is assumed to be isotropic.

$$k_c = k_m \left[\frac{1 + AB\phi}{1 - BC\phi} \right]$$

Where

$$A = 2$$

$$B = \frac{\frac{k_p}{k_m} - 1}{\frac{k_p}{k_m} + A}$$

$$C = 1 + \phi \left(\frac{1 - 0.637}{0.637^2} \right)$$

Where k_c is the effective thermal conductivity of the composite, k_m and k_p are the thermal conductivities of the matrix and the particulate respectively, ϕ is the volume fraction of the particulate and *A*, *B* and *C* are the constant whose value is given above

Table 2.1 Values of A for different shape particulate

Shape of the Particulate	Heat flow direction	A
Cube	All	2
Sphere	All	1.5

Agrawal et al. [41] derived some of the mathematical correlation for hybrid polymer composite filled with two different particulate filler material to calculate the effective thermal conductivity. These model and correlation are based on the minimum thermal resistance law and specific equivalent thermal conductivity law. These model provides the values of effective thermal conductivity of hybrid composite material which has a good agreement with the experimental effective thermal conductivity values of such hybrid composites.

$$k_c = \frac{1}{\frac{1}{k_m} - \frac{1}{k_m}\left(\frac{6\phi}{\pi}\right)^{\frac{1}{3}} + \frac{4}{\left[k\left(\frac{4\pi}{3\phi}\right)^{\frac{2}{3}} + \left(\frac{2\phi}{9\pi}\right)^{\frac{1}{3}} \times 2\pi\left(k_p - k_m\right)\right]}}$$

Agrawal et al. [27] developed heat conduction model and investigated on the enhancement of thermal conductivity of AIN/Epoxy composites and proposed a mathematical correlation between filler content and thermal conductivity of composite based on this model. By this proposed model it is seen that with the increase of the volume fraction above 40%, there is sudden increase in the effective thermal conductivity and if the volume fraction increases above the 55% then thermal conductivity of these composite turns to negative.

$$k_c = k_m \left[\frac{\dfrac{2k_m + k_p}{k_m - k_p} - 2\phi - 0.525\left(\dfrac{3k_m - 3k_p}{4k_m - 3k_p}\right) \times \phi^{\frac{10}{3}}}{\dfrac{2k_m + k_p}{k_m - k_p} + \phi - 0.525\left(\dfrac{3k_m - 3k_p}{4k_m + 4k_p}\right) \times \phi^{\frac{10}{3}}} \right]$$

2.5 Study of Composites materials filled with TiO_2

Although many polymer and its composite are found so as to use them in electronic industries as packaging material but the filler material that are found and used with polymer matrix doesn't exhibit high thermal conductivity. Some high thermal conductivity filler materials like AIN, BN, Al_2O_3 etc. are used in composite material because of their high thermal conductivity and their effect are seen on the overall effective conductivity of the composite material which found to be considerable. TiO_2 having the moderate thermal conductivity, low coefficient of thermal expansion and high electrical resistivity found to be effective as filler material. TiO_2 particles are added homogeneously ion epoxy grade composite and it is seen that flexural strength increases by 4% by using 10 wt.% filler content and 5% increment by using 20wt.% filler content, tensile modulus increase by 4% by using 10 wt.% filler content and 6% increment by using 20wt.%filler content and impact strength increase by 91% by using 10 wt.% filler content and 92% increment by using 20wt.%filler content [42].

Jyotishkumar et al. [43] found in preparation and properties of TiO_2-filled poly/epoxy Hybrid composite that incorporation of TiO_2 in the polymer matrix results in increase in the fracture toughness, impact strength and tensile properties of the polymer matrix. For filler loading 0.7wt % of TiO_2 modified ABS-epoxy hybrid composite, there is increase in the mechanical properties observed but at higher filler, mechanical properties seems to be lower than the base blend material because of the particle agglomeration in the hybrid composites. Wacharawichanant et al. [44] reported the effect of TiO_2 particle size and mixing condition on mechanical properties of polypropylene- TiO_2 and showed that increase in filler content of TiO_2 results in decrease in the tensile strength and with increase in the particle size leads to increase in the mechanical properties of the composite material. Also it is found in the result that with the mixing condition there is improvement in the distribution of TiO_2 particle which leads to increase in the mechanical properties also.

Epoxy-micro-composites and epoxy Nano-composites are prepared by taking the different volume fraction of TiO_2 and it is found that at lower volume fraction (less than 0.7 vol.%) there is increase in the fracture toughness, flexural strength and young modulus of the composite material. But as the volume fraction increases fracture toughness and flexural strength decreases while the Young modulus increases. [45]

2.2 Knowledge Gap in Earlier Work

Many of the research work is done in so many articles but despite such research work, the gap in the knowledge is seen in these researches which requires an organised and efficient research in the field of particulate filled polymer composites. Literature review mentioned above shows the following Gaps:

- ✓ So many research is done to study to enhance the thermal conductivity of the polymer matrix by addition of metal powders and so many particulate but only few works uses metal oxide as a filler or particulate material.

- ✓ Most of the studies are done experimentally on the thermal behaviour of polymer composite filled with particulate and only few focus on the development of theoretical models.

- ✓ The models developed in the past research work for the estimation of thermal conductivity of composite material doesn't provide the more approximate value of thermal conductivity when matched with the experimental value.

- ✓ Previously only the mechanical and electrical properties are modified by using TiO_2 in polymer composite material by so many investigators but for increasing the thermal conductivity of epoxy, it is used sometimes only.

The present work focuses on the effective thermal conductivity of epoxy matrix filled with TiO_2 particulates. Due to moderate thermal conductivity (12 W/m-K), high electrical resistivity, low coefficient of thermal expansion and no toxicity are the main reason to choose TiO_2 as filler material.

2.3 Objective for Present research

The objectives for following work are as follows

1) To develop a theoretical model for estimating effective thermal conductivity (k_{eff}) of polymer composites reinforced with particulate fillers.
2) Fabrication of epoxy filled with TiO_2 particulates for different volume fraction and to measure their effective thermal conductivity.
3) To study the effect of filler volume fraction and the conductivity of filler and on effective thermal conductivity of the composite system.
4) To validate the proposed mathematical correlation through experimental measurement of thermal conductivity of polymer composites fabricated with different filler concentration.

Chapter Summary

This chapter reported the following points

- ✓ A complete literature review of research works done by different investigator on distinct work of polymer composites filled with particulate filler material.
- ✓ The knowledge gap in all the earlier works.
- ✓ The objectives for Present research

The forthcoming chapter will present the various material required for fabricating the different composites.

Chapter 3

Material and Methods details

In this chapter the description is provided about the details of the materials and methods employed for fabricating and modelling the composites. The characterization of thermal behavior is also provided in this part by experimentally measuring the thermal conductivity of the sample of epoxy- TiO_2 composites with the help of appropriate instrument. It also describes the experimental methods employed to estimate the effective thermal conductivity of composite material.

3.1 MATERIALS

3.1.1 Matrix material (Epoxy)

Generally used matrix material are metals, ceramic, carbon and polymer. The most commonly used matrix material is Polymer due to its advantageous over other material like effective cost, easiness in fabrication of complex structure parts with low cost tool, stable at elevated temperature etc. Polymer matrix are present in either be in thermoplastic nature or in thermoset nature. In thermoset nature, cross-linked polymer matrix are formed by irreversible chemical transformation of resin. It has big molecular structure because of which it provides both thermal and electrical insulation. It has less viscosity, high thermal stability and high creep resistance [131]. Polyester, phenolic resin, vinyl ester and epoxy are the generally used thermoset.

In thermoplastic polymer intermolecular forces permits it to be remolded. Actually the interaction between the molecules increases with cooling and thus again restore its bulk properties. The production of these thermoplastic polymer are in one step. They can be reused and recycled. Due to their, such properties they are mostly used in the industrial application.

Teflon, polyethylene, polypropylene, polyvinyl chloride, nylon and acrylic arte the most commonly thermoplastic polymer found in market.

Epoxy are most widely used polymer matrix due to its easily availability, better mechanical and thermal properties over other polymer, good adhesion properties to different fibres, chemical resistance and better performance at higher temperature. Epoxy LY 556 resin is used as a matrix material which generally chemically belongs to epoxide family. The common name of this material is Bisphenol-A-Diglycidyl-Ether. Epoxy is chosen because it has low density ($1.1 gm/cm^3$) and it is most common used thermoset polymer and chemical resistant. It is used with its corresponding hardener HY 951. It has low thermal conductivity (0.363W/mK)

Table 3.1 Properties of Epoxy

Properties	Values
Density (gm/cc)	1.1
Thermal conductivity (W/mK)	0.363

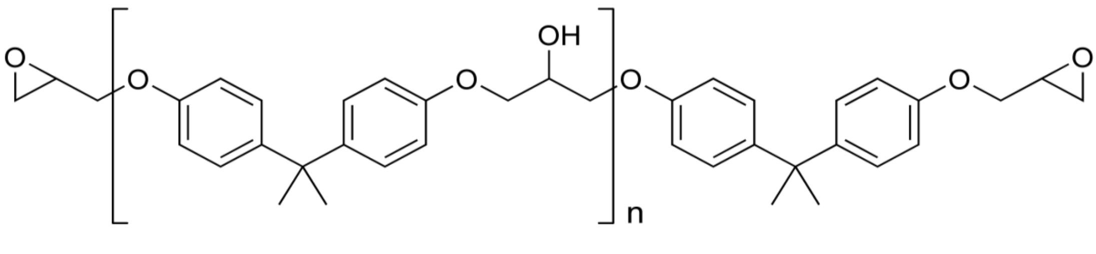

Figure 3.1 Epoxy resin chain

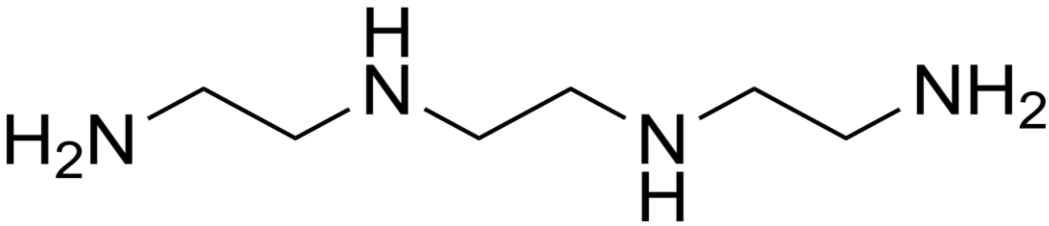

Figure 3.2 Hardener HY 951 (Tri-ethylene-tetramine)

Figure 3.3 Epoxy resin and its corresponding hardener

3.1.2 Filler material (TiO_2)

Micro-sized TiO_2 is used as a particulate filler material for the fabrication of thermal conductive PMCs in this work. It is found in the nature as anatase, brookite, and rutile in the form of oxide of titanium. Ilmenite is the main source of TiO_2. Ilmenite ore is abundant form of TiO_2 and next is Rutile. The anatase and brookite are the metastable phases and by them, they can be converted into rutile. Micro-sized titanium dioxide (TiO_2) is used as the filler material. It is preferred over other material due to Moderate thermal conductivity (12 W/m-K), Low electrical conductivity, and Low coefficient of thermal expansion

Table 3.2 Properties of TiO_2 (filler material)

Properties	Values
Density (gm/cc)	1.6
Thermal conductivity (W/mK)	0.363

Figure 3.4 TiO_2 Filler material

3.2 EXPERIMENTAL DETAILS

3.2.1 Composite Fabrication

3.2.1.1 TiO_2 filled Epoxy composites (Hand lay-up technique):-

Polymer matrix (epoxy) Composite filled with TiO_2 sample is fabricated by hand lay-up technique which is the quite old but very simple technique.

The fabrication of epoxy-TiO_2 composites is done in following manner

Polymer matrix (epoxy) Composite filled with TiO_2 sample is fabricated by hand lay-up technique. Epoxy LY 556 resin cured at Low temperature taken as matrix material along with the hardener (HY951). These both are mixed in the ratio of 10:1 by weight as mentioned. Epoxy having low density (1.1 gm/cc) and low magnitude thermal conductivity (0.363 W/m-K). Micro-sized particulates of TiO_2 are mixed in this epoxy resin to make the composites. This dough (epoxy filled with titanium oxide) is then slowly pour off into the cylindrical glass to get the composite specimen having disc shape The castings are kept at room temperature for around 26 hours to sustain complete polymerization, then from the glass moulds samples are taken for further testing of thermal conductivity of this fabricated sample.

Table 3.3 Set of Epoxy-TiO_2 composites

Composition
Epoxy + 0 vol.% TiO_2
Epoxy + 6.7 vol.% TiO_2
Epoxy + 13.4 vol.% TiO_2
Epoxy + 26.8 vol.% TiO_2
Epoxy + 40.2 vol.% TiO_2

Process illustrate the fabrication of Epoxy-TiO_2 composites by hand lay-up technique is shown in figure 3.5 and figure 3.6 shows the samples of fabricated composites.

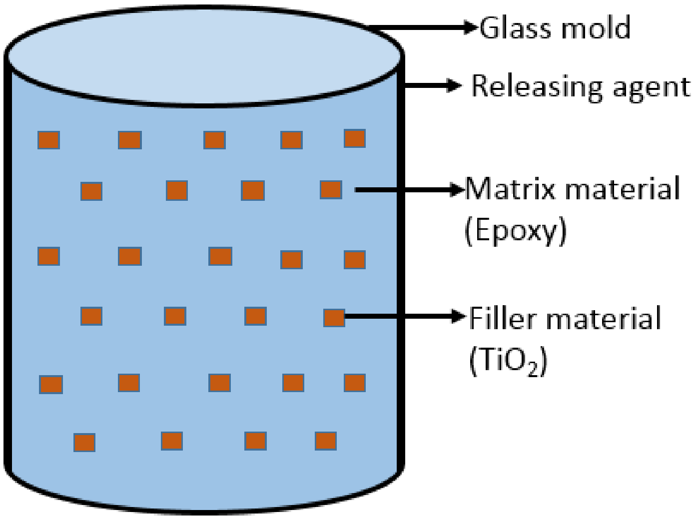

Figure 3.5 Fabrication of TiO_2 filled Epoxy composites

Figure 3.6 Fabricated composite for filler volume fraction of 6.7%, 13.4%, 26.8% and 40.2%

Experimental measurement

Unitherm™ 2022 tester is used to measure the effective thermal conductivity (k_{eff}) of epoxy-TiO_2 composite for these volume fraction as per ASTM E-1530. In this tester the sample is held in between the upper and lower plate held at some temperature gradient under a uniform compressive load. The lower plate contains the transducer. The heat flows from the upper plate to lower plate. The temperature gradient is recorded by sensors accompanied with the output heat flow from the heat transducer. These results or output from the tester are used to calculate the thermal conductivity of samples by Fourier's law of heat conduction.

$$q = kA\frac{\Delta T}{\Delta x}$$

Figure 3.7 Unitherm™ Model 2022 Tester

3.2.2 Thermal Conductivity measurement: Development of Analytical model

Depending upon the position of particulate within the polymer composites, the model developed would be vary. In the figure given below shows a 3-D view of heat transfer element of polymer composite filled with particulate. In this heat transfer element it is assumed that the cubical particulate filler material are uniformly distributed and located within the cubical polymer matrix as face-centred cubic arrangement. The reason to choose face-centred cubic arrangement is that the in the fcc arrangement the particulates are efficiently closely packed in the lattice In the given figure a heat transfer model is taken out. The particulate are assumed to be cubical and the whole element is divided into section. In each section R_i represents the corresponding thermal resistance where *i* denotes the number of the corresponding layer. Thus in this model the thermal resistors are connected in series to the direction of heat flow.

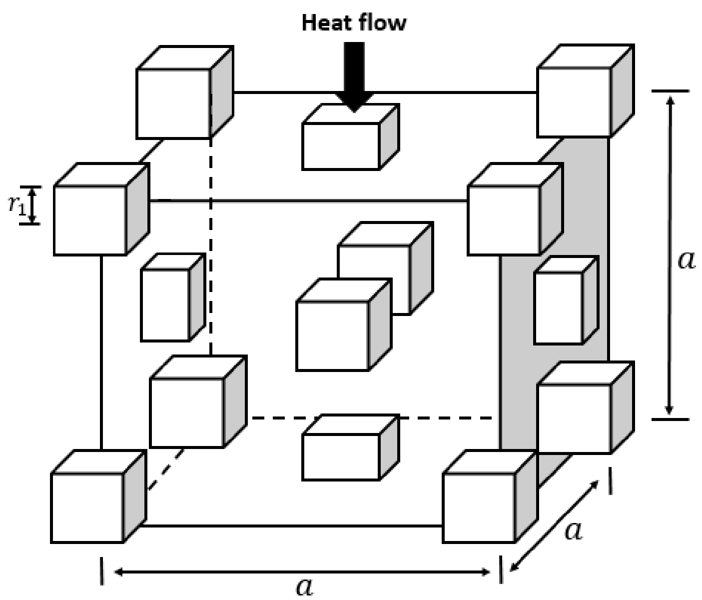

Figure 3.8 A 3-D heat transfer element in a polymer composite filled with cubical particulates in fcc arrangement

According to Fourier's law of heat conduction, the amount of heat transfer (q) through the heat transfer element is written as equation:

$$q = k_{eff} a^2 \frac{\Delta T}{a}$$

Or $q = k_{eff} a \Delta T$

And the thermal resistance of this heat transfer element is

$$R_{total} = \frac{1}{k_{eff} a}$$

Heat transfer through the matrix and particulate in the whole matrix can be expressed by Fourier's law heat conduction as

For matrix

$$q_m = k_m A_m \frac{dT}{dy}$$

For Particulate

$$q_p = k_p A_p \frac{dT}{dy}$$

Where k_m and k_p represents the thermal conductivities of the matrix and particulate respectively and A_m and A_p are the corresponding cross sectional area of the matrix and particulate respectively.

Thus the total heat transfer (q_t) through the whole heat transfer element is

$$q_t = q_m + q_p = k_m A_m \frac{dT}{dy} + k_p A_p \frac{dT}{dy} = k' A \frac{dT}{dy}$$

A represents the heat transfer elements cross sectional area

thus

$$k' = k_m \frac{A_m}{A} + k_p \frac{A_p}{A}$$

2D diagram of this heat transfer element is shown below

For No. of cube (particulate) within the matrix = 4

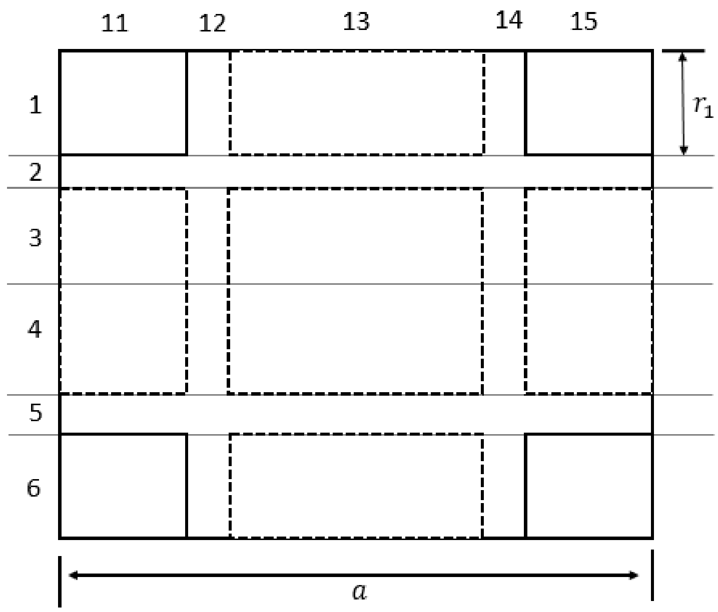

Figure 3.9 Side view of heat transfer element

Here 1, 2, 3, 4, 5 and 6 represents the horizontal layer and 11, 12, 13, 14 and 15 represents the horizontal layer upto only first horizontal layer.

Let R_1, R_2, R_3, R_4, R_5 and R_6 represents the thermal resistance of the corresponding first layer in horizontal direction

Consider the first layer then

$$\frac{1}{R_1} = \frac{1}{R_{11}} + \frac{1}{R_{12}} + \frac{1}{R_{13}} + \frac{1}{R_{14}} + \frac{1}{R_{15}}$$

Now For 11 layer

Area of matrix $A_m = ar_1 - 2r_1^2$

and Area of particulate

$A_p = 2r_1^2$ then

Effective thermal conductivity of 11 layer is

$$k'_{11} = k_m(ar_1 - 2r_1^2) + k_p \times 2r_1^2$$

$$k'_{11} = 2r_1^2(k_p - k_m) + k_m ar_1$$

Thus thermal resistance for 11 layer is

$$R_{11} = \frac{r_1}{2r_1^2(k_p - k_m) + k_m ar_1}$$

Now For 13 layer

Area of matrix $2ar_1 - 4r_1^2$

and Area of particulate

$A_p = 2r_1^2$ then

Effective thermal conductivity of 11 layer is

$$k'_{13} = k_m(2ar_1 - 4r_1^2) + k_p \times 4r_1^2$$

$$k'_{13} = 4r_1^2(k_p - k_m) + 2k_m ar_1$$

Thus thermal resistance for 13 layer is

$$R_{13} = \frac{r_1}{4r_1^2(k_p - k_m) + 2k_m ar_1}$$

And for 12 layer the thermal resistance is

$$R_{12} = \frac{2r_1}{k_m a(a - 4r_1)}$$

Now by the geometry it is seen that $R_{11} = R_{15}$ and $R_{12} = R_{14}$

Thus R_1 is calculated as

$$\frac{1}{R_1} = \frac{2}{R_{11}} + \frac{2}{R_{12}} + \frac{1}{R_{13}}$$

Now putting the above equation in this equation, we get

$$\frac{1}{R_1} = \frac{4r_1^2(k_p - k_m) + 2k_m ar_1 + k_m a(a - 4r_1) + 4r_1^2(k_p - k_m) + 2k_m ar_1}{r_1}$$

$$\frac{1}{R_1} = \frac{8r_1^{\,2}(k_p - k_m) + k_m a^2}{r_1}$$

thus

$$R_1 = \frac{r_1}{8r_1^{\,2}(k_p - k_m) + k_m a^2}$$

R_2 can be calculated as

$$R_2 = \frac{a - 4r_1}{2k_m a^2}$$

Now total thermal resistance is

$$R_{total} = R_1 + R_2 + R_3 + R_4 + R_5 + R_6$$

Also by observing the geometry it is seen that

$$R_1 = R_3 = R_4 = R_6$$

and $R_2 = R_5$

thus $R_{total} = 4R_1 + 2R_2$

$$R_{total} = \frac{4r_1}{8r_1^{\,2}(k_p - k_m) + k_m a^2} + \frac{a - 4r_1}{k_m a^2}$$

and finally the effective thermal conductivity is

$$k_{eff} = k_{total} = \frac{a}{R_{total} \times a^2} = \frac{1}{R_{total} \times a}$$

$$k_{eff} = \frac{1}{R_{total} \times a}$$

Chapter-4

RESULTS AND DISCUSSION

In this chapter the results for effective thermal conductivity of epoxy-TiO_2 composites are presented. The results from all the theoretical model proposed by different author for the estimation of effective thermal conductivity are described in this chapter.

4.1 Effective thermal conductivity of Epoxy- TiO_2 composites

4.1.1 Calculation and comparison of effective thermal conductivity of Epoxy-TiO_2 composites among proposed model, co-existing model and experimental measurement

Effective thermal conductivity of Epoxy-TiO_2 composites is calculated by the equation provided above for different loading of volume fraction of particulate. These value are calculated for distinct volume fraction of 0%, 6.7%, 13.4%, 26.8% and 40.2%. The values obtained are 0.363W/m-K, 0.6185W/m-K, 0.8269W/m-K, 1.406 W/m-K and 2.72 respectively for these volume fraction For volume fraction of 13.4% and 26.8%, these value of k_{eff} of obtained from this proposed model, are shown in figure 4.1 and figure 4.2 along with the magnitude of k_{eff} for previous models and correlation. Each of these figures fig. 4.1 and fig. 4.2 gives the comparison of theoretical value of k_{eff} among proposed model, all previous model and experimentally measured values.Also the percentage error between the theoretical models and experimental value is provide in the table. These comparative results provided in the table are illustrated graphically for their comparison with experimental values. The observation are carried out with the help of these graphs and discussion is done based on these results for the validation of this model.

Table 4.1 Comparison of k_{eff} for distinct model for 13.4 % of filler volume fraction (k_{exp} = 0.739W/m-K)

Co-existing Model	k_{eff} (W/m-K)	Percentage error (%)
Proposed Model	0.82609	11.89
Maxwell Model	0.5150	30.31
Rules of mixture Model	0.4172	43.54
Lewis & Nielson Model	0.5265	28.75
Rayleigh Model	0.40259	45.52

Table 4.2 Comparison of k_{eff} for distinct model for 26.8 % of filler volume fraction (k_{exp} = 1.232W/m-K)

Co-existing Model	k_{eff} (W/m-K)	Percentage error (%)
Proposed Model	1.406	14.12
Maxwell Model	0.71469	41.89
Rules of mixture Model	0.4904	60.19
Lewis & Nielson Model	0.7806	36.63
Rayleigh Model	0.70422	42.83

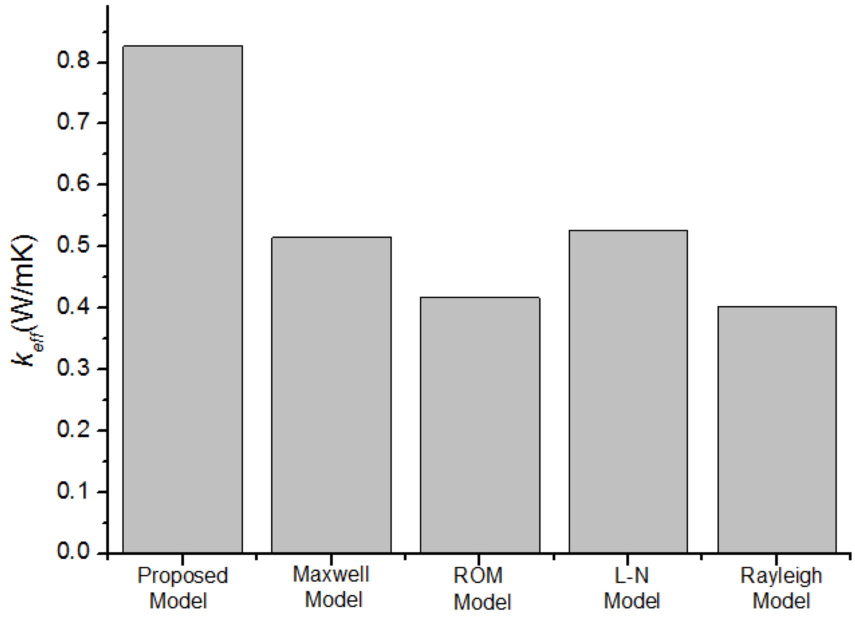

Figure 4.1 Comparision of thermal conductivity of Epoxy-TiO_2 composite for distinct model at 13.4% volume fraction of TiO_2

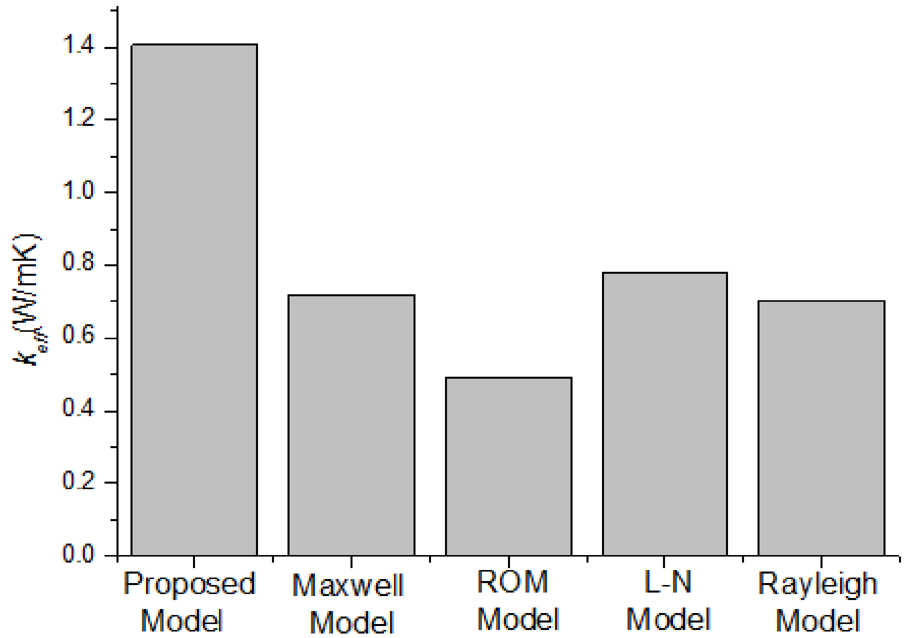

Figure 4.2 Comparision of thermal conductivity of Epoxy-TiO_2 composite for distinct model 26.8% volume fraction of TiO_2

4.1.2 Comparison of effective thermal conductivity of Epoxy-TiO_2 composites for different volume fraction of filler material

The effect of filler content is seen through the figure 4.3 which discus about the variation of thermal conductivity of epoxy-TiO_2 composites with the variation of volume fraction of TiO_2 particulate

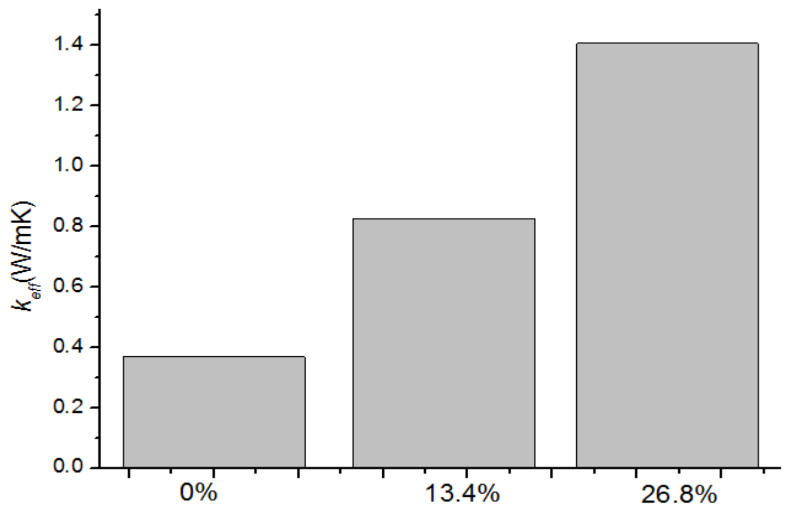

Figure 4.3 Comparision of effective thermal conductivity of Epoxy-TiO_2 composite at different volume fraction

4.1.3 Comparison among the theoretical and experimental value of effective thermal conductivity of Epoxy-TiO_2 composites for different volume fraction of filler material

The Comparison among the theoretical and experimental value of effective thermal conductivity shown in the Table 4.3 and figure 4.4 for different volume fraction of TiO_2 particulate. The gap between these theoretical and experimental values shows the percentage error in the theoretical value to the experimental value of effective thermal conductivity of epoxy- TiO_2 composite.

Table 4.3 Comparison of k_{eff} through theoretical and experimental measurements at different volume fraction

Volume fraction (ϕ)	**Effective thermal conductivity (W/m-K)**	
	Theoretical value	Experimental value
0%	0.363	0.3963
6.7%	0.6185	0.5572
13.4%	0.82609	0.739
26.8%	1.406	1.232
40.2%	2.72	2.37

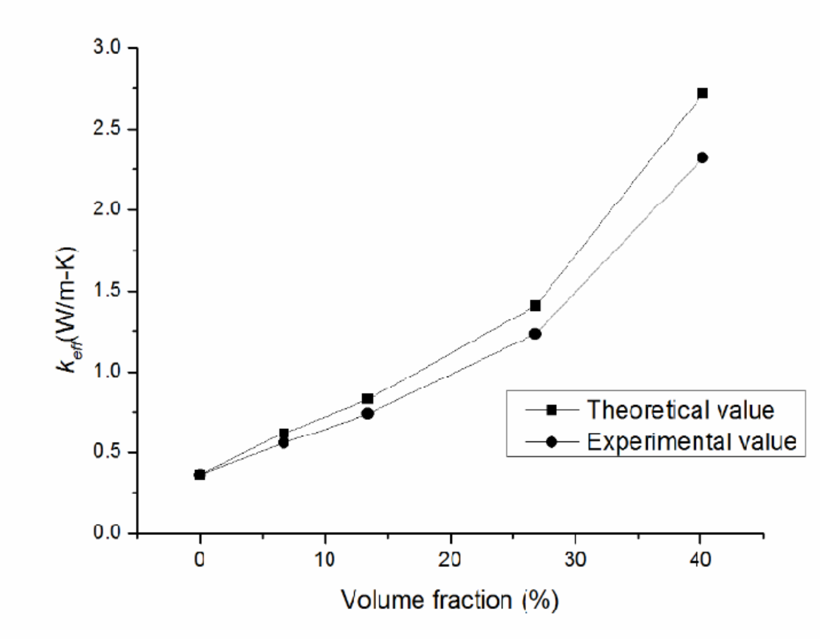

Figure 4.4 Comparision of theoretical and experimental k_{eff} of Epoxy-TiO_2 composite

4.2 Observations

These results gives the following observation:

1) With increase in the TiO_2 content in the composite the effective thermal conductivity improves quite notably. This indicates that the incorporation of micro sized TiO_2 helps in the enhancing the heat conduction capability.
2) The mathematical correlation proposed by previous investigators are found to be underrating the value of effective thermal conductivity and the deviation from the measured value are large.
3) The correlation proposed in this work provide value of effective thermal conductivity for different volume fraction of particulates are found to be in good consensus with the measured value as illustrated in figure 4.4.
4) There is increase in the gap in-between the theoretical and experimental lines of magnitude of effective thermal conductivity.

Chapter – 7

Summary and Conclusions

The work discussed in this thesis contains following parts:

1. In the first one, material used for the fabrication and the experiment related to these fabrication are described. Test performed for the calculation of thermal conductivity of the fabricated composite epoxy filled with TiO_2 particulate and its related result are also described in this part.
2. In the second part description about the development of an analytical heat conduction model is given and the mathematical correlation based on this model has been proposed for the calculation of effective thermal conductivity of epoxy composite filled with uniformly distributed, cubical shaped, micro-sized TiO_2 particles.
3. In the third part, this model and correlations are validated through comparison among co-existing model and experiment.

7.1 Summary of Work

For the microelectronic industry, the performance of the material used for its application like electronic packaging is decided by its mechanical, physical and thermal behavior which is required for the selection of that material. Sometimes the mechanical properties are sustain by so many materials but estimation of thermal behavior become mandatory for the fabrication of new material. The present work shows the information of such materials and the applicable material for such application like epoxy-TiO_2 composite is fabricated by hand lay-up technique in the laboratory for different filler volume fraction. An analytical model for the calculation of thermal conductivity of this composite and its corresponding mathematical correlation are proposed. Also for estimation of thermal conductivity, test has been performed on it. It is seen that by incorporation of TiO_2 filler particles in the epoxy matrix, the thermal conductivity of composite material increases also with the rise in the volume fraction of TiO_2 particulate, effective thermal conductivity increases. The proposed model is validated through comparison of magnitude of effective thermal conductivity among proposed model, co-existing model and experimental measurement.

7.2 Conclusions

The present analytical and experimental work has led to the following conclusion:

1) Successful fabrication of epoxy composites reinforced with micro-sized TiO_2 particulates is possible by hand lay-up technique.
2) With addition of these TiO_2 particulates, heat conduction capability of epoxy is notably increased.
3) A mathematical correlation has been developed by taking the one-dimension heat conduction across the cube-in-cube three-dimensional physical model to estimate the effective thermal conductivity.
4) It is seen that the effective thermal conductivity of such particulates filled composites is a function of filler content and of the intrinsic properties of filler and matrix materials.
5) The correlation is validated by conducting laboratory scale measurement of effective thermal conductivity of these composites and then by comparing the test result.
6) It is found that the results obtained with the addition of 6.7%, 13.4%, 26.8% and 40.2% the thermal conductivity increases by 53%, 103%, 238% and 552% respectively.
7) It is also found that the results obtained from the proposed correlation are in very good agreement with the measured value.
8) With the increase in the filler content, the percentage error increases in the measurement of effective thermal conductivity. This increase in the error is due to the domination of increase in the thermal contact resistance over decrease in the thermal resistance of composites.

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