Study on Effective Thermal Conductivity of Copper Particle Filled Polymer Composites

A Project Report Submitted in Partial Fulfillment of the Requirements for the Degree of

B. Tech.

(Mechanical Engineering)

By

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Department of Mechanical Engineering

NATIONAL INSTITUTE OF TECHNOLOGY

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C E R T I F I C A T E

This is to certify that the work in this thesis entitled ***Study on Effective Thermal Conductivity of Copper Particle Filled Polymer Composites*** by ***Kunal K Saraf,*** has been carried out under my supervision in partial fulfillment of the requirements for the degree of **Bachelor of Technology** in ***Mechanical Engineering*** during session 2010 - 2011 in the Department of Mechanical Engineering, National Institute of Technology, Rourkela.

To the best of my knowledge, this work has not been submitted to any other University/Institute for the award of any degree or diploma.

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CONTENTS

ABSTRACT

Guarded heat flow meter test method is used to measure the thermal conductivity of Copper powder filled epoxy composites using an instrument Unitherm^{TM} Model 2022 in accordance with ASTM-E1530. In the numerical study, the finite-element package ANSYS is used to calculate the conductivity of the composites. Three-dimensional spheres-in-cube lattice array models are used to simulate the microstructure of composite materials for various filler concentrations. This study reveals that the incorporation of copper particulates results in enhancement of thermal conductivity of epoxy resin and thereby improves its heat transfer capability. The experimentally measured conductivity values are compared with the numerically calculated ones and it is found that the values obtained for various composite models using finite element method (FEM) are in reasonable agreement with the experimental values.

Key Words: *Polymer Composite, Ceramic Powder Reinforcement, Thermal Conductivity, FEM*

LIST OF FIGURES AND TABLES

Figures:

Tables:

Chapter 1
Introduction

Chapter 1

INTRODUCTION

Composite Materials:

Composites are combinations of two materials in which one of the materials, called the reinforcing phase, is in the form of fiber sheets or particles and are embedded in the other material called the matrix phase. The primary functions of the matrix are to transfer stresses between the reinforcing fibers/particles and to protect them from mechanical and/or environmental damage whereas the presence of fibers/particles in a composite improves its mechanical properties such as strength, stiffness etc. A composite is therefore a synergistic combination of two or more micro-constituents that differ in physical form and chemical composition and which are insoluble in each other. The main objective is to take advantage of the superior properties of both materials without compromising on the weakness of either. Several light weight and high strength applications have successfully substituted the traditional materials by Composite materials. High strength-to-weight ratio, high tensile strength at elevated temperatures, high creep resistance and high toughness are the reasons why composites are selected for such applications. Typically, in a composite, the reinforcing materials are strong with low densities while the matrix is usually a ductile or tough material. If the composite is designed and fabricated correctly it combines the strength of the reinforcement with the toughness of the matrix to achieve a combination of desirable properties not available in any single conventional material. The strength of the composites depends primarily on the amount, arrangement and type of fiber and /or particle reinforcement in the resin. [1]

Types of Composite Materials:

Broadly, composite materials can be classified into three groups on the basis of matrix material. They are:

a) Metal Matrix Composites (MMC)

b) Ceramic Matrix Composites (CMC)

c) Polymer Matrix Composites (PMC)

a) Metal Matrix Composites:

Higher specific modulus, higher specific strength, better properties at elevated temperatures and lower coefficient of thermal expansion are the advantages of metal Matrix Composites over monolithic metals. Because of these attributes metal matrix composites are under consideration for wide range of applications viz. combustion chamber nozzle (in rocket, space shuttle), housings, tubing, cables, heat exchangers, structural members etc.

b) Ceramic matrix Composites:

One of the main objectives in producing ceramic matrix composites is to increase the toughness. Naturally it is hoped and indeed often found that there is a concomitant improvement in strength and stiffness of ceramic matrix composites.

c) Polymer Matrix Composites:

Polymeric matrix composites are the most commonly used matrix materials. The reasons for this are two-fold. In general the mechanical properties of polymers are inadequate for many structural purposes. In particular their strength and stiffness are low compared to metals and ceramics. By reinforcing other materials with polymers these difficulties can be overcome.

Secondly high pressure and high temperature are not required in the processing of polymer matrix composites. Also simpler equipment is required for manufacturing polymer matrix composites. For this reason polymer composites developed rapidly and became popular for structural applications with no time. Polymer composites are used because overall properties of the composites are superior to those of the individual polymers. They have a greater elastic modulus than the neat polymer but are not as brittle as ceramics.

Types of polymer composites:

Broadly, polymer composites can be classified into three groups on the basis of reinforcing material. They are:

Fiber reinforced polymer (FRP)

Particle reinforced polymer (PRP)

Structural polymer composites (SPC)

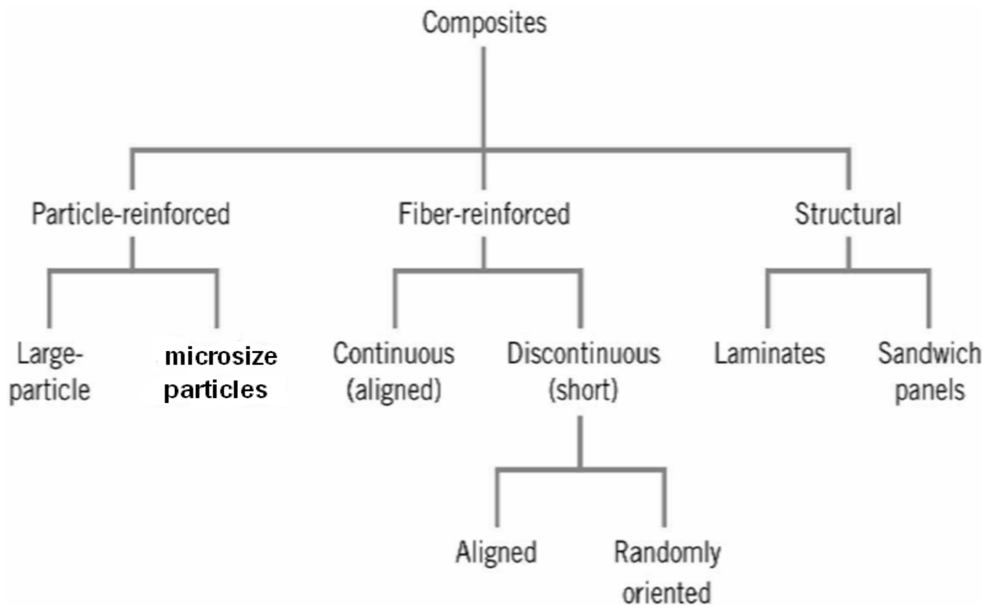

Fig. 1.1: Classification of composites based on reinforcement type

Fiber reinforced polymer:

Fibers and matrix is the main constituent of common fiber reinforced composites. Fibers are the reinforcement and the main source of strength while matrix glues all the fibers together in shape and transfers stresses between the reinforcing fibers. Loads along the longitudinal directions are carried by the fibers. Sometimes, for smoothening of the manufacturing process filler might be added to it, impact special properties to the composites and / or reduce the product cost. Common fiber reinforcing agents include asbestos, carbon/ graphite fibers, beryllium, beryllium carbide, beryllium oxide, molybdenum, Copper oxide, glass fibers, polyamide, natural fibers etc. Similarly epoxy, phenolic resin, polyester, polyurethane, vinyl ester etc. are the common matrix materials. Polyester is most widely used among these resin materials,. Epoxy, which has higher adhesion and less shrinkage than polyesters, comes in second for its high cost.

Particle reinforced polymer:

Ceramics and glasses such as small mineral particles, metal particles such as Copper and amorphous materials, including polymers and carbon black Particles used for reinforcing.

Particles are used for increasing the modulus and to decreasing the ductility of the matrix. Particles are also used for reducing the cost of the composites. Reinforcements and matrices can be common, inexpensive materials and are easily processed. High melting temp., low density, high strength, stiffness, wear resistance, and corrosion resistance are some of the useful properties of ceramics and glasses. Many ceramics are good electrical and thermal insulators. Some ceramics have special properties; some ceramics are magnetic materials; some are piezoelectric materials; and a few special ceramics are even superconductors at very low temperatures. However, ceramics and glass have one major drawback: they are brittle. An example of particle – reinforced composites is an automobile tire, which has carbon black particles in a matrix of poly-isobutylene elastomeric polymer.

Structural Polymer Composites:

These are laminar composites composed of layers of materials held together by matrix. Sandwich structures also fall under this category. Over the past few decades, it has been found that polymers have replaced many of the conventional metals/materials in various applications. Because of the advantages polymers offer over conventional materials, this has been possible. The ease of processing, productivity and cost reduction are the most important advantages of using polymers. They have generated wide interest in various engineering fields, particularly in aerospace applications. New researches are underway worldwide to develop newer composites with varied combinations of fillers and fibers so that they can be usable under all operational conditions. In most of these applications, the properties of polymers are modified using fillers and fibers to suit the high strength/high modulus requirements. Fiber-reinforced polymers offer advantages over other conventional materials when specific properties are compared. That’s the reason for these composites finding applications in diverse fields from appliances to spacecraft’s.

A lot of work has been carried out on various aspects of polymer composites, but a not so many researchers have reported on the thermal conductivity modification of particulate filled polymers. In view of this, the present work is undertaken to estimate and measure the effective thermal conductivity of epoxy filled with metal powders.

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Chapter 2

Literature Review

Chapter 2

LITERATURE REVIEW

The purpose of this literature review is to provide background information on the issues to be considered in this thesis and to emphasize the relevance of the present study. This treatise embraces some related aspects of polymer composites with special reference to their thermal conductivity characteristics. The topics include brief review:

- On Particulate Reinforced Polymer Composites
- On Thermal Conductivity of Polymer Composites
- On Thermal Conductivity Models

On particulate filled polymer composites:

Hard particulate fillers consisting of ceramic or metal particles and fiber fillers made of glass are extensively being used these days to dramatically improve the mechanical properties such as wear resistance, even up to three orders of magnitude [2]. Various kinds of polymers and polymer matrix composites reinforced with metal particles have a wide range of industrial applications such as heaters, electrodes [3], composites with thermal durability at high temperature [4] etc. These engineering composites are desired due to their low density, high corrosion resistance, ease of fabrication and low cost [5-7]. Similarly for over two decades, ceramic filled polymer composites have been the subject of extensive research. The inclusion of inorganic fillers into polymers for commercial applications is primarily aimed at the cost reduction and stiffness improvement [8, 9]. Along with fiber reinforced composites, the particulate filled composites have been found to perform well in many real operational conditions. Important role in improving electrical, mechanical and thermal properties of the composites is played by silica particles when they are added into a polymer matrix to form a composite, [10, 11]. Currently, particle size is being reduced rapidly and many studies have focused on how single-particle size affects mechanical properties [12-18]. Mechanical properties of the composites have greatly been affected by the shape, size, volume fraction, and specific surface area of such added particles. In this regard, Yamamoto et al. [19] reported that the structure and shape of silica particle have significant effects on the mechanical properties such as fatigue resistance, tensile and fracture properties. Nakamura et al. [20-22] discussed the effects of size and shape of silica particle on the strength and fracture toughness based on particle-matrix adhesion and also found an increase of the flexural and tensile strength as specific surface area of particles increased.

On Thermal Conductivity of Polymer Composites

Considerable work has been reported on the subject of heat conductivity in polymers by Hansen and Ho [23], Peng et. al [24], Choy and Young [25], Tavman [26] etc. That Increment of thermal transport significantly in the direction of orientation anddecrement slightly in the direction perpendicular to the orientation is a well-known fact. But most of these studies were confined to the thermal behavior of neat polymers only and not to their composites. Reports are available in the existing literature on experimental as well asnumerical and analytical studies on thermal conductivity of some filled polymer composites [27-39]. The fillers most frequently used are Copper particles, copper particles, brass particles, short carbon fiber, carbon particles, graphite, Copper nitrides and magnetite particles. Exhaustive overview on models and methods for predicting the thermal conductivity of composite systems was first presented by Progelhof et. al [40].Nielsen model was used by Procter and Solc [41] as a prediction to investigate the thermal conductivity of several types of polymer composites filled with different fillers and confirmed its applicability. Nagai [42] found that Bruggeman model for Al2O3/epoxy system and a modified form of Bruggeman model for AlN/epoxy system are both goodprediction theories for thermal conductivity. Griesingeret. al [43] reported that thermalconductivity of low-density poly-ethylene (LDPE) increased from 0.35 W/mK for anisotropic sample, to the value of 50 W/mK for a sample with an orientation ratio of 50.The thermal and mechanical properties of copper powder filled poly-ethylene composites are found by Tavman [44] while on thermal properties such as thermal conductivity, thermal diffusivity and specific heat of metal(copper, zinc, iron, and bronze) powder filled HDPE composites in the range of filler content 0–24% by volume were investigated experimentally by Sofian et al. [45]. A moderate increase in thermal conductivity upto 16% of metal powder filler content was observed. The improvement in electrical and thermal conductivity of polymers filled with metal powders was reported by Mamunya et. al [46]. In a recent research Weidenfeller et al. [47] studied the effect of the interconnectivity of the filler particles and its important role in the thermal conductivity of the composites. They prepared PP samples with different commercially available fillers by extrusion and injection molding using various volume fractions of filler content to systematically vary density and thermal transport properties of these composites.

Surprisingly, they measured that the thermal conductivity of the PP has increased from0.27 up to 2.5W/mK with 30 vol% talc in the PP matrix, while the same matrix material containing the same volume fraction of copper particles had a thermal conductivity of only 1.25W/m-K despite the fact that thermal conductivity of copper particles have a value approximately 40 times greater than that of talc particles. Tekce et. al [48] noticed of the shape factor of fillers has a strong influence on thermal conductivity of the composite. While Kumlutas and Tavman [49] carried out a numerical and experimental study on thermal conductivity of particle filled polymer composites, the existence of a possible correlation between thermal conductivity and wear resistance of particulate filled composites were reported by Patnaik et. al reported [50].

On Thermal Conductivity Models

Many theoretical and empirical models have been proposed to predict the effective thermal conductivity of two-phase mixtures. Comprehensive review articles have discussed the applicability of many of these models [27, 51]. For a two-component composite, the simplest alternatives would be with the materials arranged in either parallel or series with respect to heat flow, which gives the upper or lower bounds of effective thermal conductivity.

For the parallel conduction model:

$$kc = (1 - \emptyset)km + \emptyset kf \quad (2.1)$$

where, kc, km, kf are the thermal conductivities of the composite, the matrix and the filler respectively and Ø is the volume fraction of filler.

For the series conduction model:

$$\frac{1}{kc} = \frac{1-\emptyset}{km} + \emptyset\frac{}{kf} \quad (2.2)$$

The correlations presented by equations (2.1) and (2.2) are derived on the basis of the Rules of Mixture (ROM). An equation relating the two-phase solid mixture thermal conductivity to the conductivity of the individual components and to two parameters was derived by Tsao [52] which describe the spatial distribution of the two phases. By assuming a parabolic distribution of the discontinuous phase in the continuous phase, Cheng and Vachon [53] obtained a solution to Tsao's [52] model that did not require knowledge ofadditional parameters. A new model for filled polymers was proposed by Agari and Uno [54],which takes into account parallel and series conduction mechanisms.

According to this model, the expression that governs the thermal conductivity of the composite is:

$$\log(kc) = \emptyset C2\log(kf) + (1-\emptyset)\log(c1\ km) \quad (2.3)$$

Where, C_1, C_2 are experimentally determined constants of order unity. C_1 shows a measure of the effect of the particles on the secondary structure of the polymer, like crystallinity and the crystal size of the polymer. While the ease of the particles to form conductive chains are shown by C_2. The more easily particles are gathered to form conductive chains, the more thermal conductivity of the particles contributes to change in thermal conductivity of the composite and C_2 becomes closer to 1. Later, the shape of the particles was taken into account and they modified the model [55]. Generally, this semi-empirical model seems to fit the experimental data well. However, for determination of the necessary constants adequate experimental data is needed for each type of composite. For an infinitely dilute composite of spherical particles, the exact expression for the effective thermal conductivity is given as:

$$\frac{k}{kc} = 1 + \frac{3(kd-kc)}{(kd+2kc)} \quad (2.4)$$

where K, K_c and K_d are thermal conductivities of composite, continuous-phase (matrix),and dispersed-phase (filler), respectively, and Ø is the volume fraction of the dispersed phase. Equation (2.4) is the well-known Maxwell equation [56] for dilute composites.

Thermal conductivity of copper powder filled polyamide composites are investigated experimentally in the range of filler content 0–30% by volume for particle shape of short fibers and 0–60% by volume for particle shapes of plates and spheres. It is seen that the experimental values for all the copper particle shapes are close to each other at low particle content as the particles are dispersed in the polyamide matrix and they are not interacting with each other [57].

Objective of the present Investigation

The objectives of this work are outlined as follows:

1. Fabrication of a new class of low cost composites using micro-sized copper powder as the reinforcing filler with an objective to improve the heat conducting properties of neat epoxy.
2. Measurement of effective thermal conductivity (K_{eff}) of these particulate filled polymer composite (with different volume fraction) experimentally.
3. Estimation of equivalent thermal conductivity of this particulate-polymer composite system using Finite Element Method (FEM).Three dimensional spheres in cube lattice array models are constructed to simulate the microstructure of the composite materials for various filler concentration.
4. The both values of effective thermal conductivity (obtained from FEM and experiment) are compared then verified and validated. Any suitable correlation between the wear coefficient and thermal conductivity is set up (if found). Because it is largely being suspected over a half decade.
5. Recommendation of these composites for suitable applications.

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Chapter 3

Materials and Methods

Chapter 3

MATERIALS AND METHODS

This chapter describes the materials and methods used for the processing of the composites under this investigation. It presents the details of the characterization and thermal conductivity tests which the composite samples are subjected to. The numerical methodology related to the determination of thermal conductivity based on finite element method is also presented in this chapter of the thesis.

MATERIALS

Matrix Material:

Epoxy LY 556 resin, chemically belonging to the 'epoxide' family is used as the matrix material. Its common name is Bisphenol-A-Diglycidyl-Ether. The low temperature curing epoxy resin (Araldite LY 556) and the corresponding hardener (HY 951) are mixed in a ratio of 10:1 by weight as recommended. The epoxy resin and the hardener are supplied by Ciba Geigy India Ltd. Epoxy is chosen primarily because it happens to the most commonly used polymer and because of its insulating nature (low value of thermal conductivity, about 0.363 W/m-K).

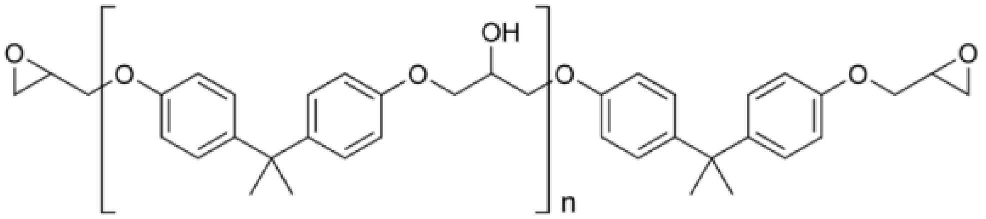

Fig. - 3.1 Epoxy Chain

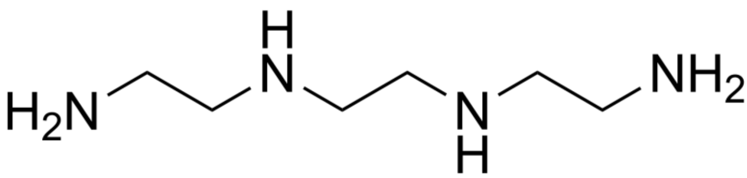

Fig. – 3.2 Triethelene tetra amine (Hardener)

Filler Material (Copper Powder):

It is a shiny dark brown metal. Copper is remarkable for the metal's high thermal conductivity and for its ability to resist corrosion .Copper and its alloys are being used in various civilizations over two millennium. Its excellent ion solubility and ease of powder production makes it a great filler material. Also it has been traditionally used in electronic packaging and electrical material. Hence for our purpose copper was selected as the filler material.

Fig 3.3 copper Power

Composite Fabrication:

The low temperature curing epoxy resin (LY 556) and corresponding hardener (HY951) are mixed in a ratio of 10:1 by weight as per recommendation. To prepare the composites, copper powder with average size 100-200µm are reinforced in epoxy resin (density 1.1gm/cc). The glass tubes coated with wax and uniform thin film of silicone-releasing agent. Then the dough (epoxy filled with Cu powder) is slowly decanted into the glass tube. Conventional hand-lay-up technique was used to cast the composite in glass tubes so as to get disk type specimens (dia. 25 mm, thickness 5 mm). Composites of four different compositions (with 0.4, 1.4, 3.34 and 6.5vol % of PWD respectively) are made. The curing time was approximately 24 hours are the dough was left for this much of time after which the tubes are broken and samples are released. Specimens of suitable dimension are cut using a diamond cutter for further characterization and thermal conductivity test.

Samples	Composition
1	Epoxy + 0 vol% (0 wt %) Filler
2	Epoxy + 0.4 vol% (3.1 wt %) Filler
3	Epoxy + 1.4vol% (10 wt %) Filler
4	Epoxy + 3.34vol% (22 wt %) Filler
5	Epoxy + 6.5vol % (48 wt %) Filler

Table 3.1: List of particulate filled composites fabricated by hand-lay-up technique

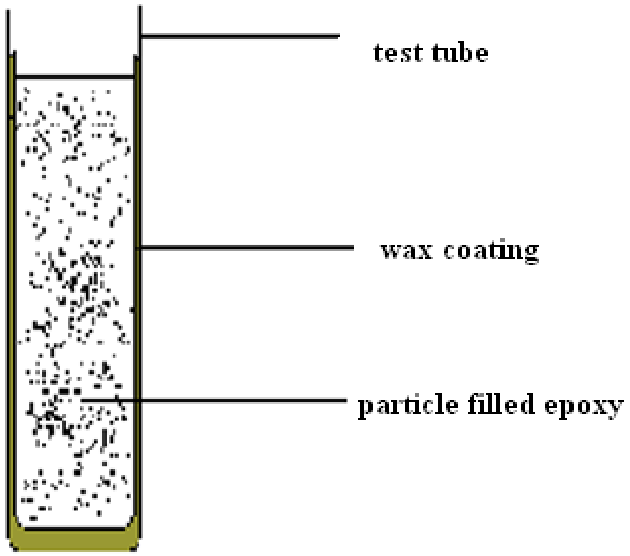

Fig. 3.4 Preparation of particulate filled composites by hand-lay-up technique

THERMAL CONDUCTIVITY CHARACTERIZATION

Experimental Determination of Thermal Conductivity:

Unitherm™ Model 2022 is used to measure thermal conductivity of a variety of materials. These include polymers, ceramics, composites, glasses, rubbers, some metals and other materials of low to medium thermal conductivity. Only a relatively small test sample is required. Non-solids, such as pastes or liquids can be tested using special containers. Thin films can also be tested accurately using a multi-layer technique.

The tests are in accordance with **ASTM E-1530** Standard.

Operating principle of Unitherm-TM 2022:

A sample of the material is held under a uniform compressive load between two polished surfaces, each controlled at a different temperature. The lower surface is part of a calibrated heat flow transducer. The heat flows from the upper surface, to the lower surface, through the sample, so that an axial temperature gradient is established in the stack. After reaching thermal equilibrium, the temperature difference across the sample is measured along with the output from the heat flow transducer. These values and the sample thickness are then used to calculate the thermal conductivity. The temperature drop through the sample is measured with temperature sensors in the highly conductive metal surface layers on either side of the sample.

Fig. 3.2 Determination of Thermal Conductivity Using Unitherm™ Model 2022

By definition thermal conductivity means "The material property that describes the rate at which heat flows with in a body for a given temperature change." For one-dimensional heat conduction the formula can be given as equation 3.1:

$$Q = \frac{KA(T1-T2)}{x} \qquad (3.1)$$

Where Q is the heat flux (W), K is the thermal conductivity (W/m-K), A is the cross sectional area (m_2) T_1-T_2 is the difference in temperature (K), x is the thickness of the sample (m). The thermal resistance of a sample can be given as:

$$R = \frac{T1-T2}{(Q/A)} \qquad (3.2)$$

Where, R is the resistance of the sample between hot and cold surfaces (m_2-K/W).

FromEquations 3.1 and 3.2 we can derive that

$$K = \frac{x}{R} \qquad (3.3)$$

In Unitherm 2022, the heat flux transducer measures the Q value and between the upper plate and lower plate the temperature difference can be obtained. Thus the thermal resistance can be calculated between the upper and lower surfaces. The thermal conductivity of the samples can be calculated using the input value of thickness and taking the known cross sectional area.

Numerical Analysis: Concept of Finite Element Method (FEM) and ANSYS:

The Finite Element Method (FEM), originally introduced by Turner et al. [59] in 1956, is a powerful computational technique for approximate solutions to a variety of "real-world" engineering problems having complex domains subjected to general boundary conditions. The physical phenomenon in various engineering disciplines has been designed or modeled using FEM. A physical phenomenon usually occurs in a continuum of matter (solid, liquid, or gas) involving several field variables. The field variables vary from point to point, thus possessing an infinite number of solutions in the domain.

The decomposition of the domain into a finite number of sub domains (elements) is relied on the basis of FEM for which the systematic approximate solution is constructed by applying the variation or weighted residual methods. In effect, the FEM problem is reduced to that of a finite number of unknowns by dividing the domain into elements and expressing the unknown field variable in terms of the assumed approximating functions within each element. These functions (also called interpolation functions) are defined in terms of the values of the field variables at specific points, referred to as nodes. Nodes are usually located along the element boundaries and they connect adjacent elements. The ability to discredit the irregular domains with finite elements makes the method a valuable and practical analysis tool for the solution of boundary, initial and Eigen value problems arising in various engineering disciplines.

A large class of engineering problems involving stress analysis, heat transfer, fluid flow etc. is thus solved using FEM. ANSYS is general-purpose finite-element modeling package for numerically solving a wide variety of mechanical problems that include static/dynamic, structural analysis (both linear and nonlinear), heat transfer, and fluid problems, as well as acoustic and electromagnetic problems.

Basic Steps in FEM:

The finite element method involves the following steps.

First, the governing differential equation of the problem is converted into an integral form.

There are two techniques to achieve this:

(i) Variational Technique

(ii) Weighted Residual Technique.

In variational technique, the calculus of variation is used to obtain the integral form corresponding to the given differential equation. The solution of the problem is obtained by minimizing the integral. For structural mechanics problems, the integral form turns out to be the expression for the total potential energy of the structure. In weighted residual technique, the integral form is constructed as a weighted integral of the governing differential equation where the weight functions are known and arbitrary except that they satisfy certain boundary conditions. This integral form is often modified using the divergence theorem to reduce the continuity requirement of the solution. The solution of the problem is obtained by initializing the integral to zero. For structural mechanics problems, if the weight function is considered as the virtual displacement, then the integral form becomes the expression of the virtual work of the structure.

In the second step, the domain of the problem is divided into a number of parts, called as elements. For one-dimensional (1-D) problems, the elements are nothing but line segments having only length and no shape. For problems of higher dimensions, the elements have both the shape and size. For two-dimensional (2D) or axi-symmetric problems, the elements used are triangles, rectangles and quadrilateral having straight or curved boundaries. When the domain boundary is curved, curved sided elements are good choice. For three-dimensional (3-D) problems, the shapes used are tetrahedron and parallelepiped having straight or curved surfaces. Division of the domain into elements is called a mesh.

In this step, over a typical element, a suitable approximation is chosen for the primary variable of the problem using interpolation functions (also called as shape functions) and the unknown values of the primary variable at some pre-selected points of the element, called as the nodes. Usually polynomials are chosen as the shape functions. For 1-D elements, there are at least 2 nodes placed at the endpoints. Additional nodes are placed in the interior of the element. For 2-D and 3-D elements, the nodes are placed at the vertices (minimum 3 nodes for triangles, minimum 4 nodes for rectangles, quadrilaterals and tetrahedral and minimum 8 nodes for parallelepiped shaped elements). Additional nodes are placed either on the boundaries or in the interior. The values of the primary variable at the nodes are called as the degrees of freedom.

The expression for the primary variable must contain a complete set of polynomials (i.e., infinite terms) to get the exact solution or if it contains only the finite number of terms, then the number of elements must be infinite. In either case, it results into an infinite set of algebraic equations. A finite number of elements and an expression with finite number of terms are used to make the problem tractable. An approximate solution is thus obtained. (Therefore, the expression for the primary variable chosen to obtain an approximate solution is called an approximation). The accuracy of the approximate solution, however, can be improved either by increasing the number of terms in the approximation or the number of elements.

In the fourth step, the approximation for the primary variable is substituted into the integral form. If the integral form is of variational type, it is minimized to get the algebraic equations for the unknown nodal values of the primary variable. If the integral form is of the weighted residual type, it is set to zero to obtain the algebraic equations. In each case, the algebraic equations are obtained element wise first (called as the element equations) and then they are assembled over all the elements to obtain the algebraic equations for the whole domain (called as the global equations). In this step, the algebraic equations are modified to take care of the boundary conditions on the primary variable. The modified algebraic equations are solved to find the nodal values of the primary variable.

In the last step, the post-processing of the solution is done. That is, first the secondary variables of the problem are calculated from the solution. Then, the nodal values of the primary and secondary variables are used to construct their graphical variation over the domain either in the form of graphs (for 1-D problems) or 2-D/3-D contours as the case may be.

Advantages of the finite element method over other numerical methods are as follows:

 The method can be used for any irregular-shaped domain and all types of boundary conditions.

 Domains consisting of more than one material can be easily analyzed.

 Accuracy of the solution can be improved either by proper refinement of the mesh or by choosing approximation of higher degree polynomials.

 The algebraic equations can be easily generated and solved on a computer. In fact, a general purpose code can be developed for the analysis of a large class of problems.

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Chapter 4
Results and Discussion

Chapter 4

RESULTS AND DISCUSSION

PHYSICAL CHARACTERIZATION

Density and void fraction:

The theoretical density of composite materials in terms of weight fraction can easily be obtained as for the following equations given by Agarwal and Broutman [58].

$$\rho ct = \frac{1}{\left\{\left(\frac{Wf}{\rho f}\right)+\left(\frac{Wm}{\rho m}\right)\right\}} \qquad (4.1)$$

Where, *W* and *ρ* represent the weight fraction and density respectively. The suffix *f, m* and *ct* stand for the fiber, matrix and the composite materials respectively.

In case of hybrid composites, consisting of three components namely matrix, fiber and particulate filler, the modified form of the expression for the density of the composite can be written as:

$$\rho ct = 1/\left\{\left(\frac{Wf}{\rho f}\right) + \left(\frac{Wm}{\rho m}\right) + \left(\frac{Wp}{\rho p}\right)\right\} \qquad (4.2)$$

Where the suffix *p'*indicates the particulate filler materials.

Samples	Composition (for Cu filled epoxy)	Density of the composite
1	Epoxy + 0 vol% (0 wt %) Filler	1.1
2	Epoxy + 0.4 vol% (3.1 wt %) Filler	1.131
3	Epoxy + 1.4vol% (10 wt %) Filler	1.210
4	Epoxy + 3.34vol% (22 wt %) Filler	1.367
5	Epoxy + 6.5vol % (48 wt %) Filler	1.611

Table 4.1 Density of the composites under this study

THERMAL CONDUCTIVITY CHARACTERIZATION

Description of the problem

For functional design and application of composite materials the determination of effective properties of composite materials is of paramount importance. Microstructure of the composite is the one of the important factors that influence the effective properties and can be controlled to an appreciable extent. Here, microstructure means the shape, size distribution, spatial distribution and orientation distribution of the reinforcing inclusion in the matrix. Although most composite possess inclusion of random distributions, great insight of the effect of microstructure on the effective properties can be gained from the investigation of composites with periodic structure. Because of the high degree of symmetry embedded in the system, periodic structures can be more easily analyzed.

Thermal analysis is carried out for the conductive heat transfer through the composite body using the finite-element program ANSYS. In order to make a thermal analysis, three-dimensional physical models with spheres-in-a-cube lattice array have been used to simulate the microstructure of composite materials for four different filler concentrations. Furthermore, the effective thermal conductivities of these epoxy composites filled with copper dust up to about 6.5% by volume is numerically determined using ANSYS.

Assumptions

In the analysis of the ideal case it will be assumed that

1. The composites are macroscopically homogeneous

2. Locally both the matrix and filler are homogeneous and isotropic

3. The thermal contact resistance between the filler and the matrix is negligible.

4. The composite lamina is free of voids

5. The problem is based on 3D physical model

6. The filler are arranged in a square periodic array/uniformly distributed in matrix.

Numerical Analysis

In the numerical analysis of the heat conduction problem, the temperatures at the nodes along the surfaces ABCD is prescribed as T_1 (=100^0C) and the convective heat transfer coefficient of ambient is assumed to be 2.5 W/m^2-K at ambient temperature of 27°C. The heat flow direction and the boundary conditions are shown in Fig. 4.1. The other surfaces parallel to the direction of the heat flow are all assumed adiabatic. The temperatures at the nodes in the interior region and on the adiabatic boundaries are unknown. These temperatures are obtained with the help of finite-element program package ANSYS.

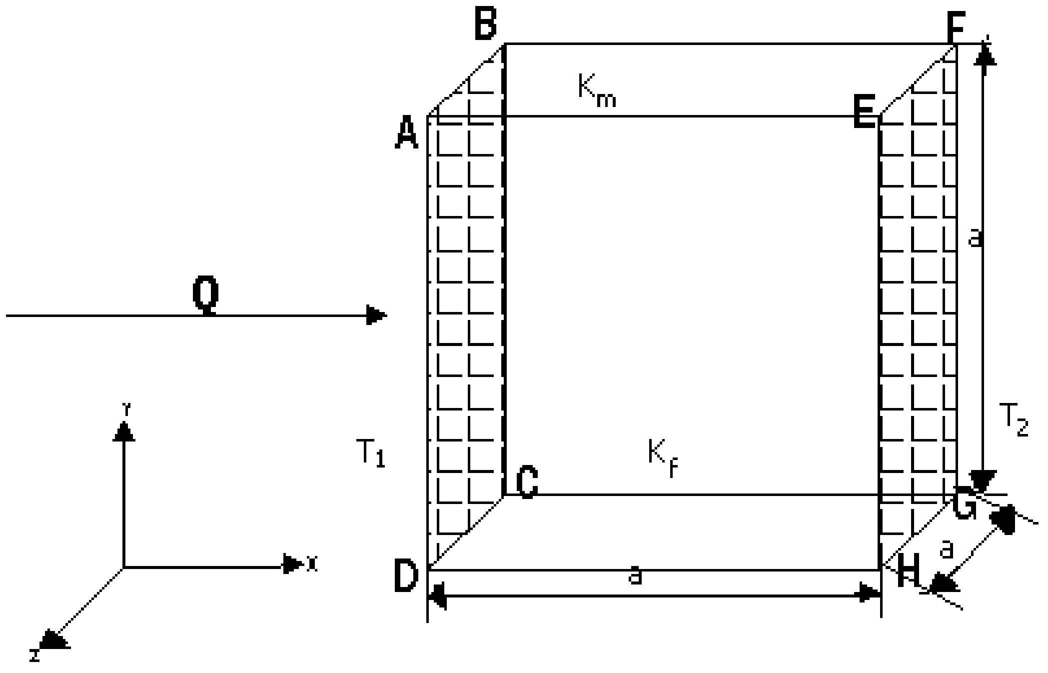


Fig.4. 1 Boundary conditions

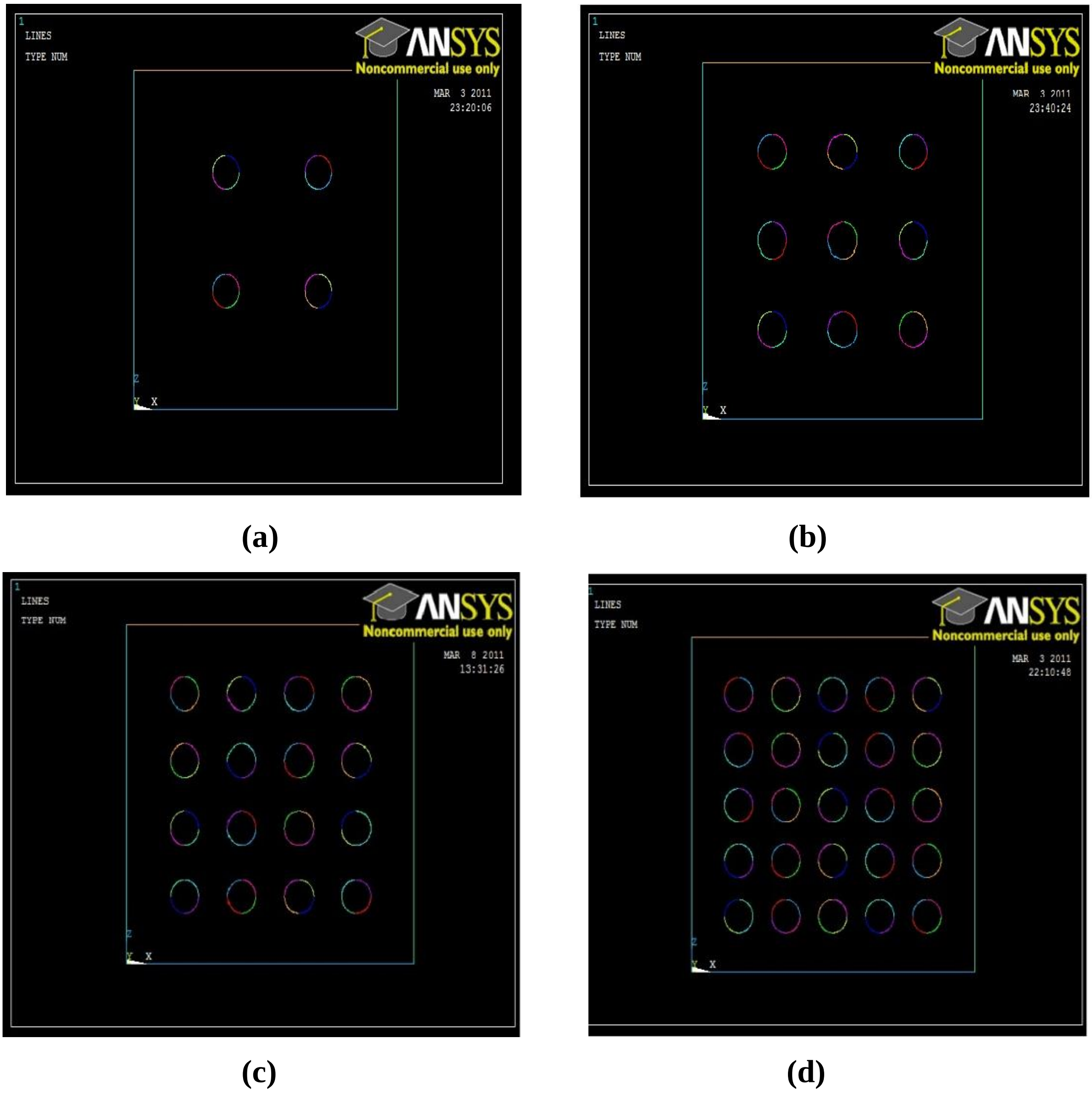
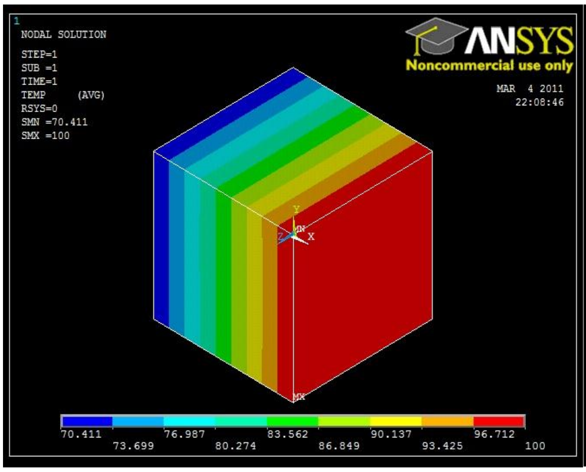
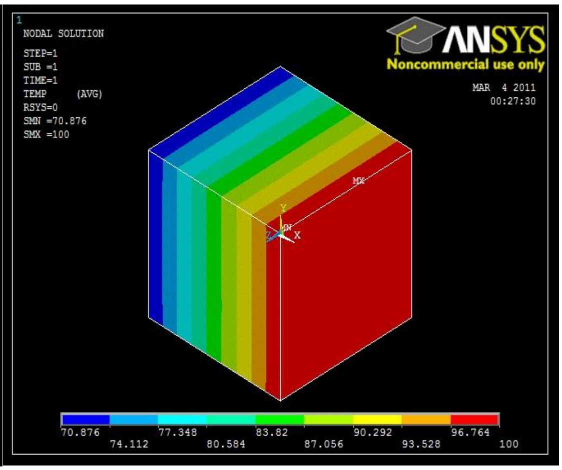


Fig. 4.2 Typical 3-D spheres-in-cube models with particle concentration of Copper powder (a) 0.4 vol% (b) 1.4 vol% (c) 3.34 (d) 6.5vol% respectively.



(a)



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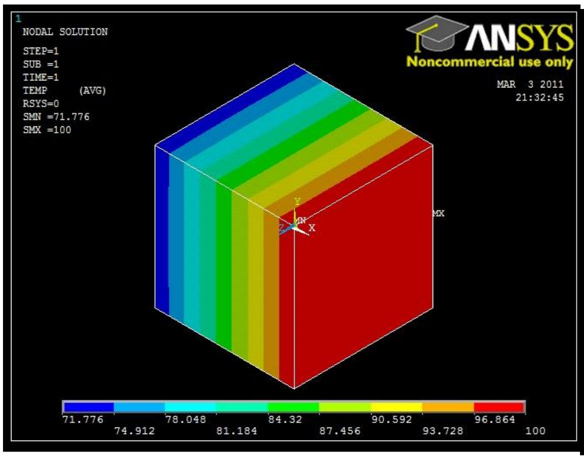

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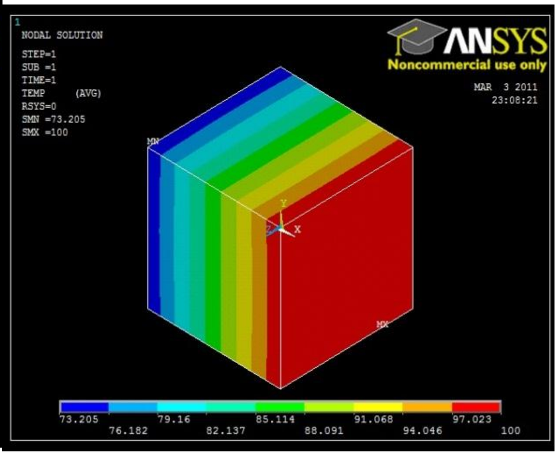

(d)

Fig. 4.3 Temperature profiles for Copper filled epoxy composites with particle concentration of (a) 0.4 vol% (b) 1.4 vol% (c) 3.34 (d) 6.5 vol% respectively

Sample	Cu Content (vol %)	Effective thermal conductivity of composites K_{eff}(W/m-K)	
		FEM simulated value (Spheres-in-cube Model)	Experimentally measured value
1	0	-	0.363
2	0.4	0.3673	0.364
3	1.4	0.3767	0.369
4	3.34	0.3966	0.385
5	6.5	0.4311	0.425

Table 4.2 Keff values for Epoxy/Cu composites obtained from FEM and Experiment

Composite Sample	Cu Content (Vol. %)	Percentage errors associated with FEM results w.r.t. the experimental value (%)
1	0.4	0.90
2	1.4	2.08
3	3.34	3.01
4	6.5	1.43

Table 4.3 Percentage errors associated with the FEM simulated values with respect to the measured values (for Cu filled epoxy composites)

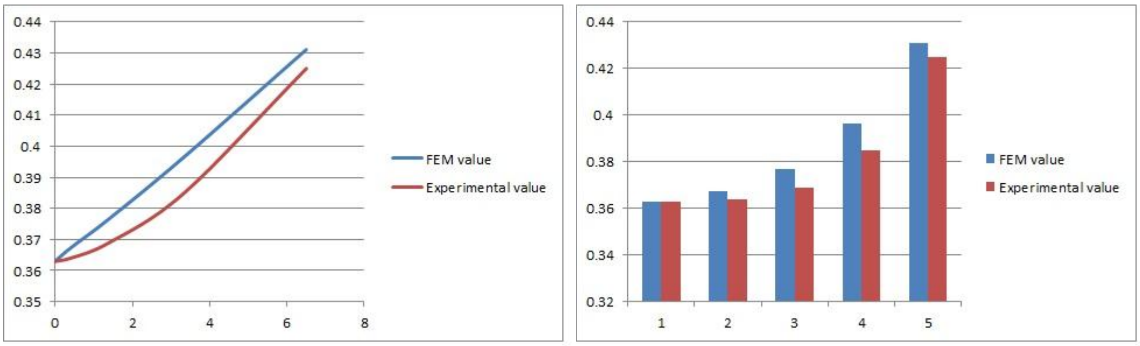

Fig 4.4 Graphs for effective thermal conductivity of copper filled composite

Thermal conductivities of epoxy composites filled with Copper particles to 6.5% by volume are numerically estimated by using the spheres-in-cube model and the numerical results are compared with the experimental results and also with some of the existing theoretical and empirical models The temperature profiles obtained from FEM analysis for the composites with particulate concentrations of 0.4, 1.4, 3.34 and 6.5 vol % are presented in Figures 4.3a - 4.3d respectively.

This study shows that finite element method can be gainfully employed to determine effective thermal conductivity of these composite with different amount of filler content. The value of equivalent thermal conductivity obtained for various composite models using FEM are in reasonable agreement with the experimental values for a wide range of filler contents from about 0.4 vol.% to 6.5 vol.%. Incorporation of Cu results in enhancement of thermal conductivity of epoxy resin. With addition of 6.5 vol. % of Cu, the thermal conductivity improves by about 18.5 % with respect to neat epoxy resin.

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Chapter 5

Conclusions

Chapter 5

CONCLUSIONS AND SCOPE FOR FUTURE WORK

Conclusions

This numerical and experimental investigation on thermal conductivity of Copper filled epoxy composites have led to the following specific conclusions:

- Successful fabrication of epoxy based composites filled with micro-sized Cuby hand-lay-up technique is possible.
- Finite element method can be gainfully employed to determine effective thermal conductivity of these composite with different amount of filler content.
- The value of equivalent thermal conductivity obtained for various composite models using FEM are in reasonable agreement with the experimental values for a wide range of filler contents from about 0.4 vol.% to 6.5 vol.%.
- Incorporation of Cu results in enhancement of thermal conductivity of epoxy resin. With addition of 6.5 vol. % of Cu, the thermal conductivity improves by about 18.5 % with respect to neat epoxy resin.
- These new class of Cu filled epoxy composites can be used for applications such as electronic packages, encapsulations, die (chip) attach, thermal grease, thermal interface material and electrical cable insulation.

Scope for future work

This work leaves a wide scope for future investigators to explore many other aspects of thermal behavior of particulate filled composites. Some recommendations for future research include:

- Effect of filler shape and size on thermal conductivity of the composites
- Exploration of new fillers for development of thermal insulation materials

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Publications

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Study on Thermal Conductivity of Metal Particle Filled Polymer Composites

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Abstract

Guarded heat flow meter test method is used to measure the thermal conductivity of metal (aluminium and copper) powder filled epoxy composites using an instrument Unitherm™ Model 2022 in accordance with ASTM-E1530. In the numerical study, the finite-element package ANSYS is used to calculate the conductivity of the composites. Three-dimensional spheres-in-cube lattice array models are used to simulate the microstructure of composite materials for various filler concentrations. This study reveals that the incorporation of metal particulates results in enhancement of thermal conductivity of epoxy resin and thereby improves its heat transfer capability. The experimentally measured conductivity values are compared with the numerically calculated ones and also with the already existing theoretical and empirical models. It is found that the values obtained for various composite models using finite element method (FEM) are in reasonable agreement with the experimental values.

Key Words: *Polymer Composite, Metal Powder Reinforcement, Thermal Conductivity, FEM*

Introduction

Hard particulate fillers consisting of ceramic or metal particles and fiber fillers made of glass are being used these days to dramatically improve the mechanical properties such as wear resistance. Various kinds of polymers and polymer matrix composites reinforced with metal particles have a wide range of industrial applications such as heaters, electrodes, composites with thermal durability at high temperature, etc. Such particulate filled polymers with higher thermal conductivities than the unfilled ones are becoming a more important area of study because of the wide range of applications, e.g., in electronic packaging and in applications with decreasing geometric dimensions and increasing output of power, like in computer chips. Considerable work has been reported on the subject of heat conductivity in polymers by Hansen and Ho (1965), Peng and Landel (1975), Choy and Young (1977), Tavman (1991) etc. The fillers most frequently used are aluminum, copper and brass particles, short carbon fiber, carbon particles, graphite, aluminum nitride and magnetite particles etc. Progelhof et.al (1976) were the first to present an exhaustive overview on models and methods for predicting the thermal conductivity of composite systems. Procter and Solc (1991) used Nielsen model

as a prediction to investigate the thermal conductivity of several types of polymer composites filled with different fillers and confirmed its applicability. Nagai (1997) found that Bruggeman model for Al_2O_3/epoxy system and a modified form of Bruggeman model for AlN/epoxy system are both good prediction theories for thermal conductivity. Griesinger et al (1997) reported that thermal conductivity of low-density poly-ethylene (LDPE) increased from 0.35 W/mK for an isotropic sample, to 50 W/mK for a sample with an orientation ratio of 50. The thermal and mechanical properties of copper powder filled poly-ethylene composites are found by Tavman (1991), while Sofian et.al (2001) investigated experimentally on thermal properties such as thermal conductivity, thermal diffusivity and specific heat of metal (copper, zinc, iron, and bronze) powder filled HDPE composites in the range of filler content 0-24% by volume. They observed a moderate increase in thermal conductivity up to 16% of metal powder filler content. Mamunya et.al (2002) also reported the improvement in electrical and thermal conductivity of polymers filled with metal powders. While Kumlutas and Tavman (2006) carried out a numerical and experimental study on thermal conductivity of particle filled polymer composites, Patnaik et.al (2010) reported the existence of a possible correlation between thermal conductivity and wear resistance of particulate filled composites. Recently Nayak et.al (2010) have reported on the modified thermal conductivity of pine wood dust filled epoxy based composites.
Against this background, the present investigation is undertaken with an objective to analyze the heat transfer through the epoxy composites filled with two conductive fillers (micro-sized aluminum and copper powder) and to evaluate the effective thermal conductivity of these composites by numerical as well as experimental methods.

Experimental details

Composite fabrication

Epoxy LY 556 resin, chemically belonging to the ‘epoxide’ family is used as the matrix material. Its common name is Bisphenol-A-Diglycidyl-Ether. The epoxy resin and the hardener are supplied by Ciba Geigy India Ltd. Epoxy is chosen primarily because it happens to be the most commonly used polymer and because of its low density (1.1 gm/cc). Aluminum and copper powder of about 100 micron mean particle size are reinforced in epoxy resin to prepare the composites. This low temperature curing epoxy resin and the corresponding hardener (HY951) are mixed in a ratio of 10:1 by weight as recommended. The dough (epoxy filled with metal powders) is then slowly decanted into the glass molds, coated beforehand with wax and a uniform thin film of silicone-releasing agent. The composites are cast in these molds so as to get disc type cylindrical specimens (dia 25 mm, thickness 5 mm). Composites of different compositions as listed in Table 1 are made. The castings are left to cure at room temperature for about 24 hours after which the glass molds are broken and samples are released.

Experimental determination of thermal conductivity

Unitherm™ Model 2022 is used to measure thermal conductivity of a variety of materials. These include polymers, ceramics, composites, glasses, rubbers, some metals, and other materials of low to medium thermal conductivity. Only a relatively small test sample is required. Non-solids, such as pastes or liquids, can be tested using special containers. Thin films can also be tested accurately using a multi-layer technique. The tests are in accordance with ASTM E-1530 standard.

Numerical Analysis: Concept of finite element method and ANSYS

The finite element method (FEM), originally introduced by Turner et al. (1956) , is a powerful computational technique for approximate solutions to a variety of ‘‘real-world” engineering problems having complex domains subjected to general boundary conditions. FEA has become an essential step in the design or modeling of a physical phenomenon in various engineering disciplines. A physical phenomenon usually occurs in a continuum of matter (solid, liquid, or gas) involving several field variables. The field variables vary from point to point, thus possessing an infinite number of solutions in the domain. The basis of FEM relies on the decomposition of the domain into a finite number of sub-domains (elements) for which the systematic approximate solution is constructed by applying the variational or weighted residual methods. In effect, FEM reduces the problem to that of a finite number of unknowns by dividing the domain into elements and by expressing the unknown field variable in terms of the assumed approximating functions within each element. These functions (also called interpolation functions) are defined in terms of the values of the field variables at specific points, referred to as nodes. Nodes are usually located along the element boundaries and they connect adjacent elements. The ability to discretize the irregular domains with finite elements makes the method a valuable and practical analysis tool for the solution of boundary, initial and eigen-value problems arising in various engineering disciplines. The FEM is a numerical procedure that can be used to obtain solutions to a large class of engineering problems involving stress analysis, heat transfer, fluid flow etc. ANSYS is general-purpose finite-element modeling package for numerically solving a wide variety of mechanical problems that include static/dynamic, structural analysis (both linear and nonlinear), heat transfer, and fluid problems, as well as acoustic and electromagnetic problems.

Results and discussion

Numerical analysis

Using the finite-element program ANSYS, thermal analysis is carried out for the conductive heat transfer through the composite body. In order to make a thermal analysis, three-dimensional physical models with spheres-in-cube array have been used to simulate the microstructure of composite materials for four different filler concentrations. Furthermore, the effective thermal conductivities of these epoxy composites filled with aluminium/copper powder up to about 3.334 % and 6.5% respectively by volume are numerically determined using ANSYS.

Description of the problem

The determination of effective properties of composites is of paramount importance for functional design and application of composite materials. One of the important factors that influence the effective properties and can be controlled to an appreciable extent is the microstructure of the composite. Here, microstructure means the shape, size distribution, spatial distribution and orientation distribution of the reinforcing inclusion in the matrix. Although most composite possess inclusion of random distributions, great insight of the effect of microstructure on the effective properties can be gained from the investigation of composites with periodic structure. System with periodic structures can be more easily analyzed because of the high degree of symmetry embedded in the system.

In the analysis of this conduction problem, the heat flow direction and the boundary conditions for the particulate-epoxy composite body are shown in Fig.1. The temperature at the nodes along the surfaces ABCD is prescribed as T1 (=100^0C) and the ambient convective heat transfer coefficient is assumed to be 25 W/m^2-K at room temperature of 27°C. The other surfaces parallel to the direction of the heat flow are all assumed adiabatic. The temperatures at the nodes in the interior region and on the other boundaries are unknown. These temperatures are obtained with the help of finite-element program package ANSYS. In this analysis it is be assumed that the composites are macroscopically homogeneous, locally both the matrix and filler are homogeneous and isotropic, the thermal contact resistance between the filler and the matrix is negligible, the composite lamina is free of voids, the problem is based on 3D physical model and the filler are arranged in a square periodic array and are uniformly distributed in matrix.

Thermal conductivities of epoxy composites filled with aluminium/copper particles up to 3.334 and 6.5 % respectively by volume are numerically estimated by using the spheres-in-cube model. Typical 3-D models showing arrangement of spherical fillers with a particle concentration of 0.4, 1.4, 3.34 and 6.5 vol % within the cube shaped matrix body is illustrated in Figs.2 (a), 2(b), 2(c) and 2(d) respectively. The temperature profiles obtained from FEM analysis for the composites (spheres-in-cube arrangement) with particulate concentrations of 0.4,1.4,3.34,6.5 vol % are presented in following figures. The numerical results are compared with the experimental results. The simulated values of effective thermal conductivity of the composites obtained by FEM analysis are presented in Table 2 along with the corresponding measured values.

The percentage errors associated with the FEM values with respect to the experimental values is given in Table 3. It is seen from this table that the errors associated with the spheres-in-cube model simulations lie in the range of 1-6 %. . Fig.5 and 6 compares the FEM simulated results of thermal conductivity with those found from experiments for aluminium and copper respectively. It also presents the variation of effective thermal conductivity as a function of the Al/Cu content in the composite. The difference between the simulated values and the measured value of conductivity may be attributed to the fact that some of the assumptions taken for the numerical analysis are not real. The shape of Al/Cu is assumed to be spherical, while in actual practice they are irregular shaped. Moreover, although the distribution of Al/Cu particulates in the matrix body is assumed to be in an arranged manner, it is actually dispersed in the epoxy almost randomly. However, it is encouraging to note that the incorporation of Al/Cu results in enhancement of thermal conductivity of epoxy resin. With addition of 3.34 vol. % of Al, the thermal conductivity improves by about 6.3 % with respect to neat epoxy resin .Similarly, with addition of 3.34% and 6.5% of Cu the thermal conductivity improves by about 9.26 and 18.67 % when compared with neat epoxy resin.

5. Conclusions

This numerical and experimental investigation on thermal conductivity of Al/Cu filled epoxy composites has led to the following specific conclusions:

1. Successful fabrication of epoxy based composites filled with micro-sized Al/Cu by hand-lay-up technique is possible.
2. Finite element method can be gainfully employed to determine effective thermal conductivity of these composite with different amount of filler content.
3. The value of equivalent thermal conductivity obtained for various composite models using FEM are in reasonable agreement with the experimental values for a wide range of filler contents from about 0.4 vol.% to 6.5 vol.%.
4. Incorporation of Al/Cu results in enhancement of thermal conductivity of epoxy resin. With addition of 3.34 vol. % of Al, the thermal conductivity improves by about 6.3 % with respect to neat epoxy resin .Similarly, with addition of 3.34% and 6.5% of Cu the thermal conductivity improves by about 9.26% and 18.67 % respectively when compared with neat epoxy resin.
5. These new class of Al/Cu filled epoxy composites can be used for applications such as electronic packages, encapsulations, die (chip) attach, thermal grease, thermal interface material and electrical cable insulation.

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TABLES AND FIGURES

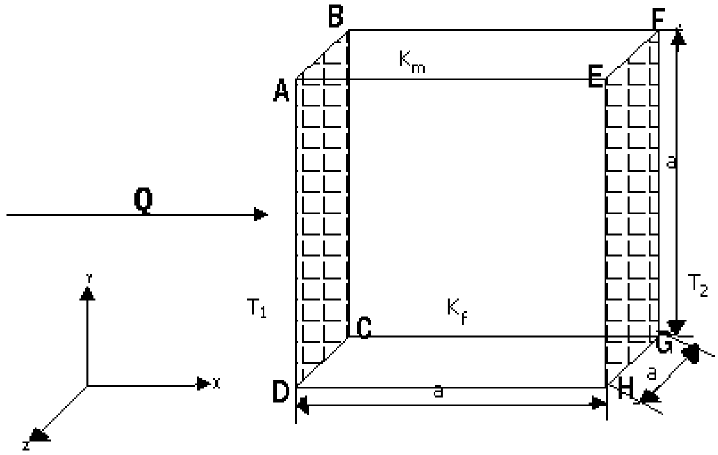
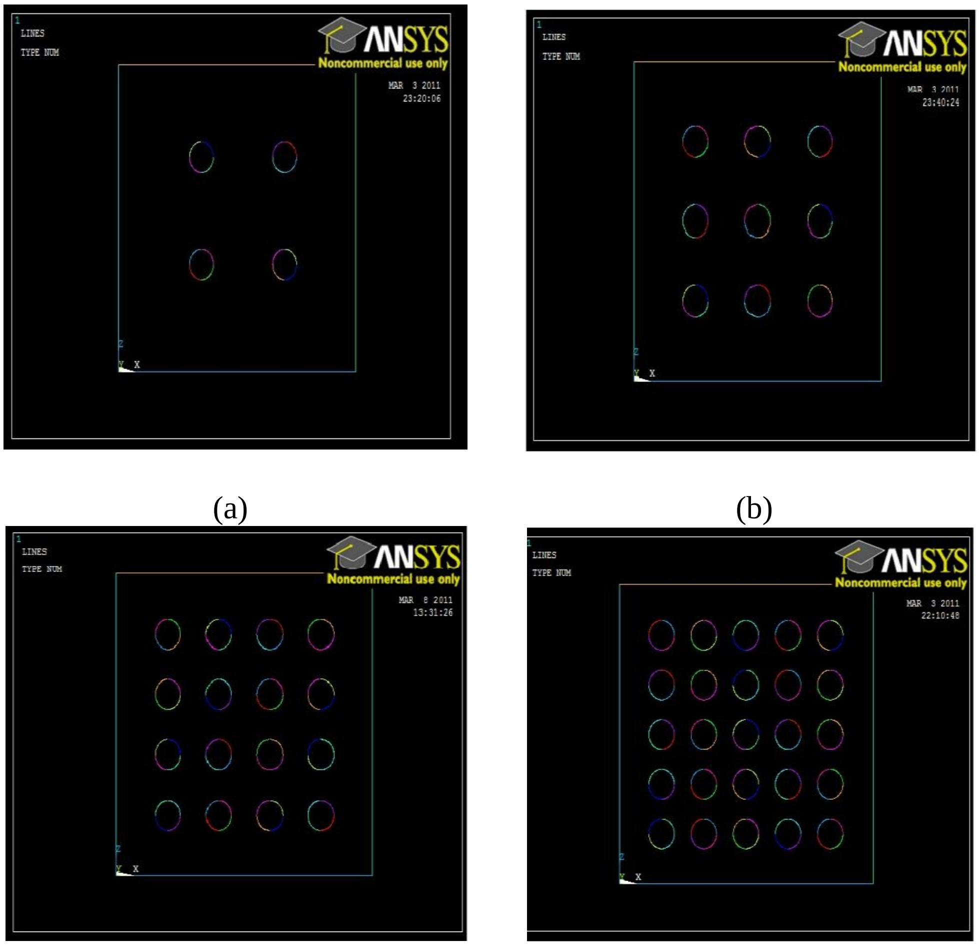


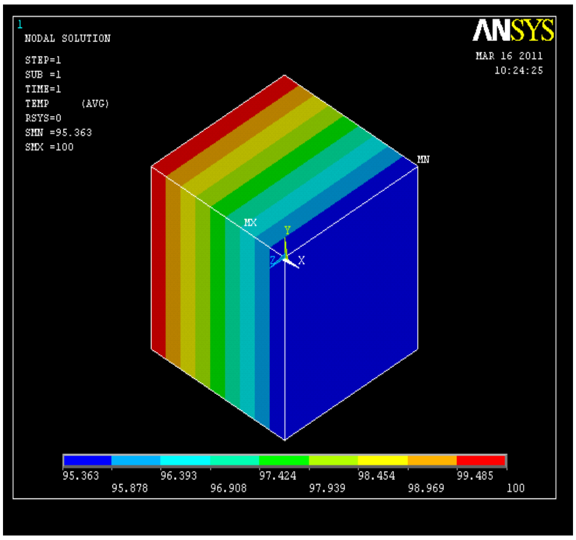
Fig. 1 Boundary conditions



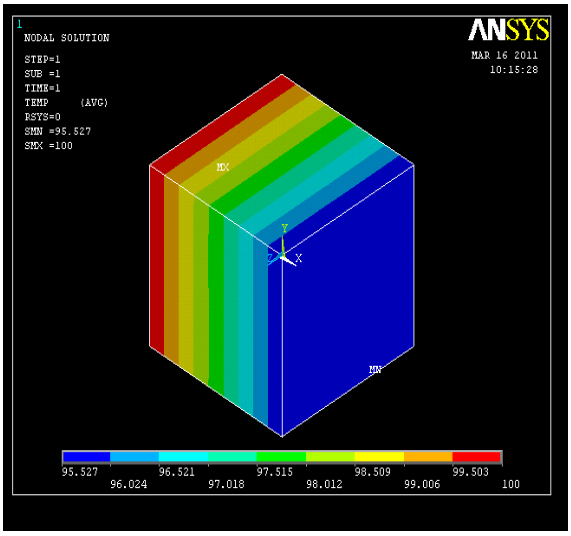
(a) (b)

(c) (d)

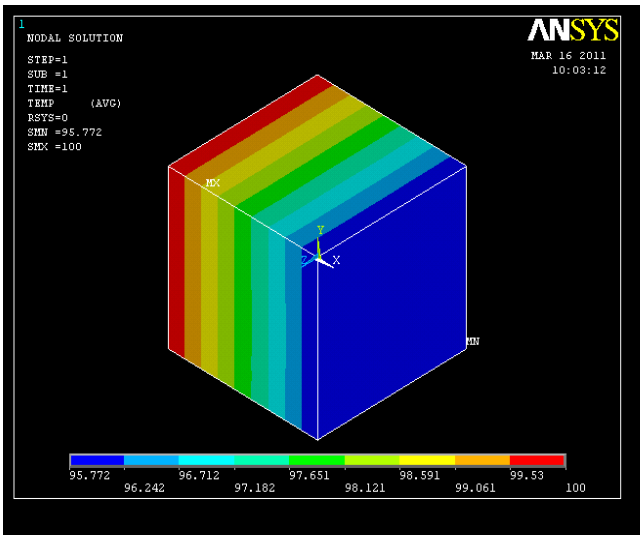
Fig. 2 Typical 3-D spheres-in-cube models with particle concentration of (a) 0.4 vol% (b) 1.4 vol% (c) 3.34 and (d) 6.5 vol% respectively



(a)

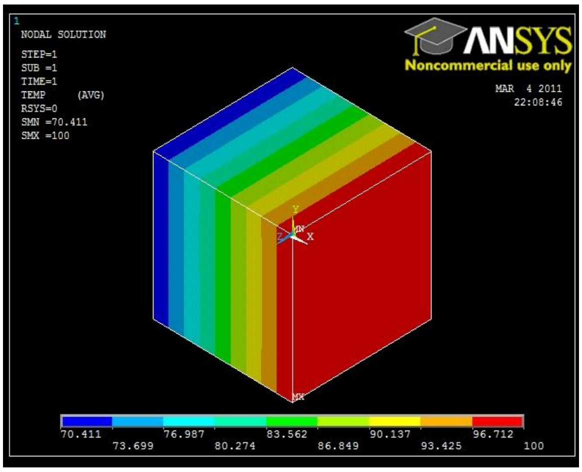


(b)

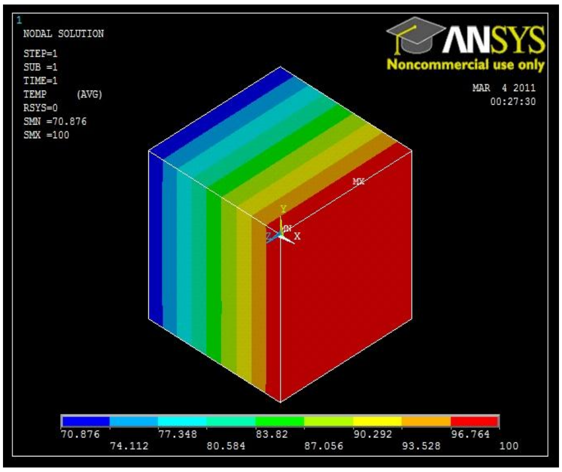


(c)

Fig. 3 Temperature profiles for aluminum filled epoxy composites with particle concentration of (a) 0.4 vol% (b) 1.4 vol% and (c) 3.34 vol% respectively



(a)



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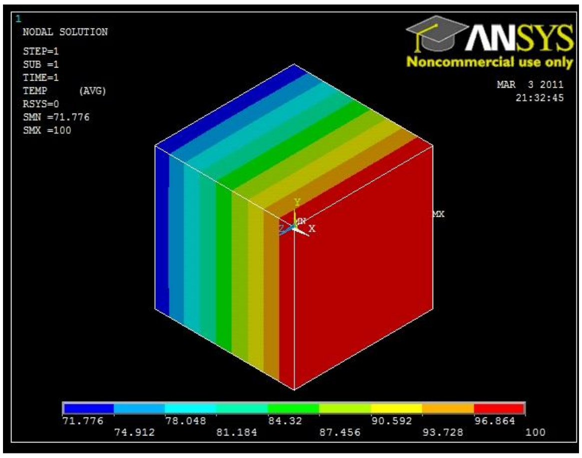
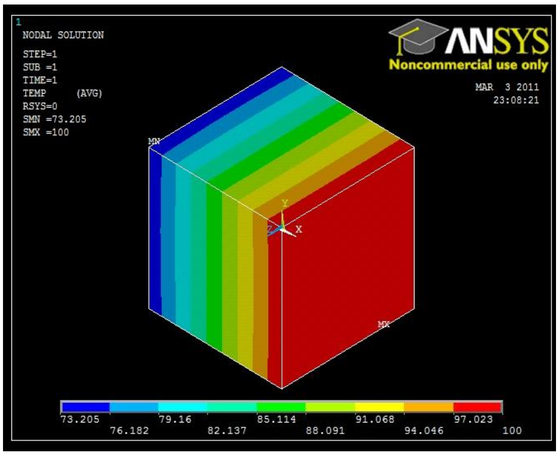


(c)



(d)

Fig. 4 Temperature profiles for copper filled epoxy composites with particle concentration of (a) 0.4 vol% (b) 1.4 vol% (c) 3.34 and (d) 6.5 vol% respectively

Samples	Composition (for Al filled epoxy)	Composition (for Cu filled epoxy)
1	Epoxy + 0 vol% (0 wt %) Filler	Epoxy + 0 vol% (0 wt %) Filler
2	Epoxy + 0.4 vol% (1.01 wt %) Filler	Epoxy + 0.4 vol% (3.1 wt %) Filler
3	Epoxy + 1.4vol% (3.4 wt %) Filler	Epoxy + 1.4vol% (10 wt %) Filler
4	Epoxy + 3.34 vol% (7.8 wt %) Filler	Epoxy + 3.34 vol% (22 wt %) Filler
5	Epoxy + 6.5 vol % (14.7 wt %) Filler	Epoxy + 6.5 vol % (48 wt %) Filler

Table 1 List of particulate filled composites fabricated by hand-lay-up technique

Sample	Al Content (vol %)	Effective thermal conductivity of composites K_{eff} (W/m-K)	
		FEM simulated value (Spheres-in-cube Model)	Experimentally measured value
1	0	-	0.363
2	0.4	0.369	0.364
3	1.4.	0.382	0.369
4	3.34	0.408	0.385

Table 2a K_{eff} values for Epoxy/Al composites obtained from FEM and Experiment

Sample	Cu Content (vol %)	Effective thermal conductivity of composites K_{eff} (W/m-K)	
		FEM simulated value (Spheres-in-cube Model)	Experimentally measured value
1	0	-	0.363
2	0.4	0.3673	0.364
3	1.4.	0.3767	0.369
4	3.34	0.3966	0.385
5	6.5	0.4311	0.425

Table 2b K_{eff} values for Epoxy/Cu composites obtained from FEM and Experiment

Composite Sample	Al Content (Vol. %)	Percentage errors associated with FEM results w.r.t. the experimental value (%)
1	0.4	1.3
2	1.4	3.5
3	3.34	5.9

Table 3a Percentage errors associated with the FEM simulated values with respect to the measured values ((for Al filled epoxy composites)

Composite Sample	Cu Content (Vol. %)	Percentage errors associated with FEM results w.r.t. the experimental value (%)
1	0.4	0.90
2	1.4	2.08
3	3.34	3.01
4	6.5	1.43

Table 3b Percentage errors associated with the FEM simulated values with respect to the measured values ((for Cu filled epoxy composites)