EFFECTIVE THERMAL CONDUCTIVITY OF EPOXY MATRIX COMPOSITES FILLED WITH TITANIA (TiO_2) POWDER

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

B. Tech.

(Mechanical Engineering)

By

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NATIONAL INSTITUTE OF TECHNOLOGY

ROURKELA

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CERTIFICATE

This is to certify that the work in this thesis entitled ***Effective Thermal Conductivity of Epoxy Matrix Composites Filled With Titania(TiO_2) Powder*** by **Sonam Agrawal**, 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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ACKNOWLEDGEMENT

I would like to express my deep sense of gratitude and respect to my supervisor Prof. Alok Satapathy for his excellent guidance, suggestions and support. I consider myself extremely lucky to be able to work under the guidance of such a dynamic personality. I am also thankful to Prof. R. K. Sahoo, H.O.D, Department of Mechanical Engineering, N.I.T., Rourkela for his constant support and encouragement.

Last but not the least, I extend my sincere thanks to all other faculty members and my friends at the Department of Mechanical Engineering, NIT, Rourkela, for their help and valuable advice in every stage for successful completion of this project report.

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C O N T E N T S

ABSTRACT

Guarded heat flow meter test method is used to measure the thermal conductivity of Titania (TiO_2) 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 titania(TiO_2) 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*

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Chapter 1
Introduction

Chapter 1

INTRODUCTION

Composite Materials:

Composites Materials are combinations of two materials in which one of the materials, called the reinforcing phase, which is in the form of fiber sheets or particles and are embedded in the other material called the matrix phase. The primary functions of this matrix are to transfer stresses between the reinforcing fibers or particles and to protect them from mechanical and environmental damage whereas the presence of fibers or 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. Our objective is to take advantage of the superior properties of both materials without compromising on the weakness of either. Composite materials have successfully substituted the conventional materials in several applications like light weight and high strength. The reasons why composites are selected for such applications are mainly dut to their high strength-to-weight ratio, high tensile strength at elevated temperatures, high creep resistance and high toughness. Typically, 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 traditional material. The strength of the composites which are made, depends primarily on the amount, arrangement and type of fiber and /or particle reinforcement in the matrix. [1]

Types of Composite Materials:

Basically, composites can be categorized 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:

These Composites have many advantages over monolithic metals like higher specific strength, higher specific modulus, better properties at elevated temperatures and lower coefficient of thermal expansion. Due to 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:

The main objective 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:

These are the most commonly used matrix material. 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. These difficulties are overcome by reinforcing other materials with polymers.

Secondly the processing of this type of matrix composites do not require high pressure and high temperature. The equipments which are required for manufacturing polymer matrix composites are simpler. For this reason polymer composites developed rapidly and soon became popular for structural applications. Polymer composites are used because overall properties of these composites are superior to those of the individual polymers. The elastic modulus is greater than that of the neat polymer but are not as brittle as ceramics.

Classification of polymer composites:

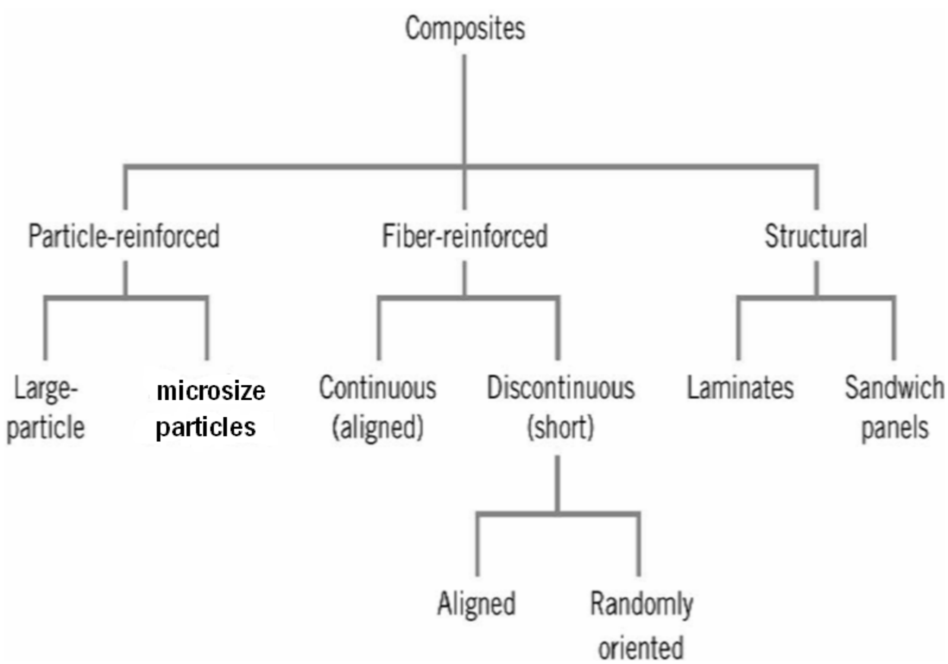

Fig. 1.1: Classification of composites based on reinforcement type

Polymer composites can be classified into following three groups on the basis of reinforcing material. They are:

(a) Fiber reinforced polymer (FRP)

(b) Particle reinforced polymer (PRP)

(c) Structural polymer composites (SPC)

(a) Fiber reinforced polymer:

The fiber reinforced composites are composed of fibers and matrix. Fibers are the reinforcing elements and the main source of strength while matrix glues all the fibers together in shape and transfers stresses between the reinforcing fibers. The fibers carry the loads along their longitudinal directions. Sometimes, filler is added to smoothen the manufacturing process and to impact special properties to the composites. These also reduce the production cost. Most commonly used agents include asbestos, carbon/ graphite fibers, beryllium, beryllium carbide, beryllium oxide, molybdenum, aluminum oxide, glass fibers, polyamide, natural fibers etc. Similarly common matrix materials include epoxy, phenolic resin, polyester, polyurethane, vinyl ester etc. Among these materials, resin and polyester are most widely used. Epoxy, which has higher adhesion and less shrinkage than polyesters, comes in second for its high cost.

(b) Particle reinforced polymer:

Particles which are used for reinforcing include ceramics and glasses such as small mineral particles, metal particles such as aluminum and amorphous materials, including polymers and carbon black. Particles are used to enhance the modulus and to decrease the ductility of the matrix. Some of the useful properties of ceramics and glasses include high melting temp., low density, high strength, stiffness; wear resistance, and corrosion resistance etc. Many ceramics are good electrical and thermal insulators. Some ceramics have special properties; some have magnetic properties; some are piezoelectric materials; and a few special ceramics are even superconductors at very low temperatures. one major drawback of ceramics and glass is their brittleness. An example of particle – reinforced composites is an automobile tyre, which has carbon black particles in a matrix of poly-isobutylene elastomeric polymer.

(c) Structural Polymer Composites:

These are the laminar composites which are composed of layers of materials held together by matrix. This category also includes sandwich structures. Over the past few decades, we find that these polymers have replaced many of the conventional materials in various applications. The most important advantages of using polymers are the ease of processing, productivity and cost reduction. The properties of polymers are modified using fillers and fibers to fulfill the high strength and 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 spacecrafts.

A lot of work has been carried out on various aspects of polymer composites, but a few 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 ceramic 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:

1) On Particulate Reinforced Polymer Composites
2) On Thermal Conductivity of Polymer Composites
3) On Thermal Conductivity Models

(1) On particulate filled polymer composites:

Hard particulate fillers consisting of ceramic or metal particles and fiber fillers made of glass are being used to improve the mechanical properties such as wear resistance [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]. 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 composites made with particulate fillers have been found to perform well in many real operational conditions. When silica particles are added into a polymer matrix to form a composite, they play an important role in improving electrical, mechanical and thermal properties of the composites [10,11]. Currently, particle size is being reduced rapidly and many studies have focused on how single-particle size affects mechanical properties [12-18]. The shape, size, volume fraction, and specific surface area of such added particles have been found to affect mechanical properties of the composites greatly. 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.

(2) 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. It is well known that thermal transport increases significantly in the direction of orientation and decreases slightly in the direction perpendicular to the orientation. Reports are available in the existing literature on experimental as well as numerical and analytical studies on thermal conductivity of some filled polymer composites [27-39]. The fillers most frequently used are aluminum particles, copper particles, brass particles, short carbon fiber, carbon particles, graphite, aluminum nitrides and magnetite particles. Progelhof et. al [40] was the first to present an exhaustive overview on models and methods for predicting the thermal conductivity of composite systems. Procter and Solc [41] 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 [42] found that Bruggeman model for Al2O3/epoxy system and a modified form of Bruggeman model for AlN/epoxy system are both good prediction theories for thermal conductivity. Griesinger et. al [43] reported that thermal conductivity of low-density poly-ethylene (LDPE) increased from 0.35 W/mK for an isotropic 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 Sofian et al. [45] 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 [46] also reported the improvement in electrical and thermal conductivity of polymers filled with metal powders. 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 from 0.27 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 copper particles have a thermal conductivity approximately 40 times greater than that of talc particles. Tekce et. al [48] noticed the strong influence of the shape factor of fillers 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, Patnaik et. al reported the existence of a possible correlation between thermal conductivity and wear resistance of particulate filled composites [50].

(3) 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:

$$kc = (1 - \emptyset)km + \emptyset kf \tag{2.1}$$

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:

$$\frac{1}{kc} = \frac{1-\emptyset}{km} + \emptyset/kf \tag{2.2}$$

The correlations presented by equations (2.1) and (2.2) are derived on the basis of the Rules of Mixture (ROM). Tsao [52] derived an equation relating the two-phase solid mixture thermal conductivity to the conductivity of the individual components and to two parameters 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 of additional parameters. Agari and Uno [54] propose a new model for filled polymers, which takes into account parallel and series conduction mechanisms.

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

$$\log(kc) = \emptyset c2 \log(kf) + (1 - \emptyset)\log(c1\, km) \tag{2.3}$$

where, C1, C2 are experimentally determined constants of order unity. C1 is a measure of the effect of the particles on the secondary structure of the polymer, like crystallinity and the crystal size of the polymer. C2 measures the ease of the particles to form conductive chains. 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 C2 becomes closer to 1. Later, they modified the model to take into account the shape of the particles [55]. Generally, this semi-empirical model seems to fit the experimental data well. However, adequate experimental data is needed for each type of composite in order to determine the necessary constants. For an infinitely dilute composite of spherical particles, the exact expression for the effective thermal conductivity is given as:

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

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

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 titania (TiO_2) powder as the reinforcing filler with an objective to improve the heat conducting properties of neat epoxy.
2. Measurement of thermal conductivity (k) of these particulate filled polymer composite experimentally.
3. Estimation of equivalent thermal conductivity of this particulate-polymer composite system using Finite Element Method (FEM).
4. Validation of the FEM analysis by measuring the thermal conductivity values experimentally.
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).

Filler Material (Titania Powder):

Titanium dioxide is the most widely used white pigment because of its brightness and very high refractive index (n = 2.7), in which it is surpassed only by a few other materials. It has a wide range of applications, from paint to sunscreen to food colouring. Its thermal conductivity and density values are 11.6 W/m.K and 4.197 gm/cc respectively. TiO_2 incorporated into outdoor building materials, such as paving stones in noxer blocks or paints, can substantially reduce concentrations of airborne pollutants such as volatile organic compounds and nitrogen oxides**.**

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 recommended. Titania(TiO_2) powder with average size 100-200 μm are reinforced in epoxy resin (density 1.1 gm/cc) to prepare the composites. The dough (epoxy filled with TiO_2 powder) is then slowly decanted into the glass tubes, coated beforehand with wax and uniform thin film of silicone-releasing agent. The composites are cast by conventional hand-lay-up technique in glass tubes so as to get cylindrical specimens (dia. 9 mm, length 120 mm). Composites of four different compositions (with 0.4, 1.4, 3.34 and 6.5 vol % of Titania(TiO_2) respectively) are made. The castings are left to cure at room temperature for about 24 hours after which the tubes are broken and samples are released. Specimens of suitable dimension are cut using a diamond cutter for further physical characterization and thermal conductivity test.

Samples	Composition (for TiO_2 filled epoxy)
1	Epoxy + 0 vol% (0 wt %) Filler
2	Epoxy + 0.4188 vol% (1.579 wt %) Filler
3	Epoxy + 1.413vol% (5.18 wt %) Filler
4	Epoxy + 3.34 vol% (11.64 wt %) Filler
5	Epoxy + 6.5 vol % (22.084 wt %) Filler

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

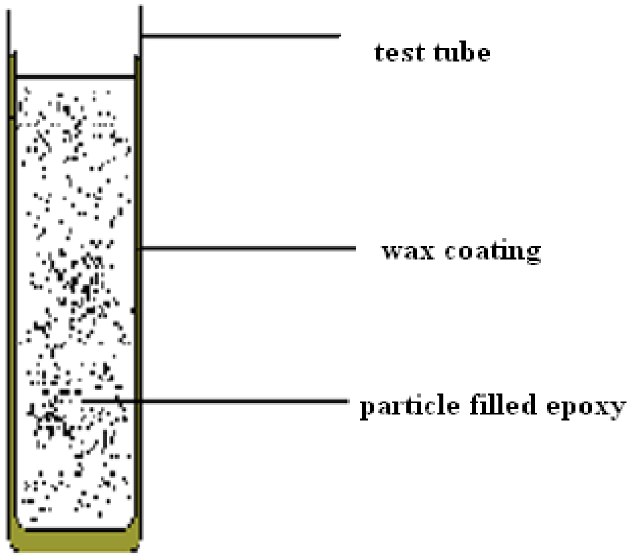

Fig. 3.1 Preparation of particulate filled composites by hand-lay-up technique

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 UnithermTM 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, through the sample, to the lower surface, establishing an axial temperature gradient 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.

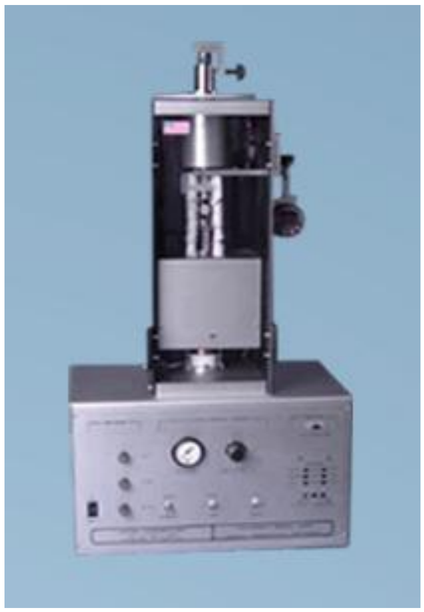

Fig. 3.2 Determination of Thermal Conductivity Using Unitherm™ Model 2022

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:

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

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). From Equations 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 the temperature difference can be obtained between the upper plate and lower plate. Thus the thermal resistance can be calculated between the upper and lower surfaces. Giving the input value of thickness and taking the known cross sectional area, the thermal conductivity of the samples can be calculated using Equation 3.3.

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

The Finite Element Method (FEM), originally introduced by Turner et al. [58] 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. FEM 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 subdomains (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 thus 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.

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. This integral needs to be minimized to obtain the solution of the problem. 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. To reduce the continuity requirement of the solution, this integral form is often modified using the divergence theorem. This integral form is set to zero to obtain the solution of the problem. 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. Curved sided elements are good choice when the domain boundary is curved. 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.

To get the exact solution, the expression for the primary variable must contain a complete set of polynomials (i.e., infinite terms) 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. To make the problem tractable, only a finite number of elements and an expression with only finite number of terms are used. Then, we get only an approximate solution. (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 canbe 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 [56].

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

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	Density(in g/cc)
1	Epoxy + 0 vol% (0 wt %) Filler	**1.1**
2	Epoxy + 0.4188 vol% (1.579 wt %) Filler	1.11
3	Epoxy + 1.413vol% (5.18 wt %) Filler	1.14
4	Epoxy + 3.34 vol% (11.64 wt %) Filler	1.20
5	Epoxy + 6.5 vol % (22.084 wt %) Filler	1.30

Table 4.1 Density of the composites under this study

THERMAL CONDUCTIVITY CHARACTERIZATION

Description of the problem

The determination of effective properties of composite materials 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.

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-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 titania 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 T1 (=100^{0}C) and the convective heat transfer coefficient of ambient is assumed to be 2.5 W/m2-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.

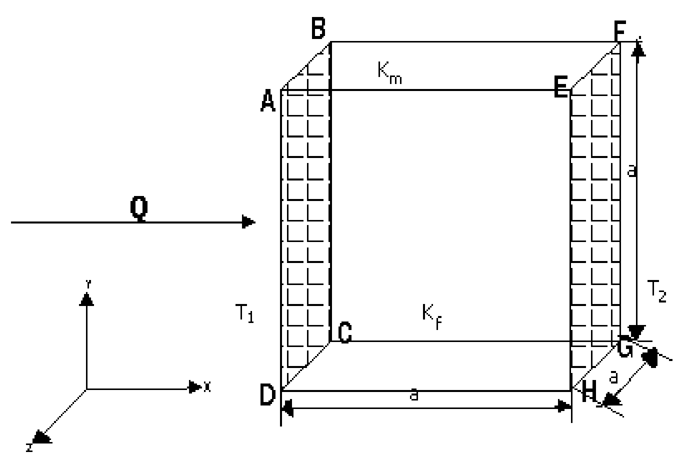
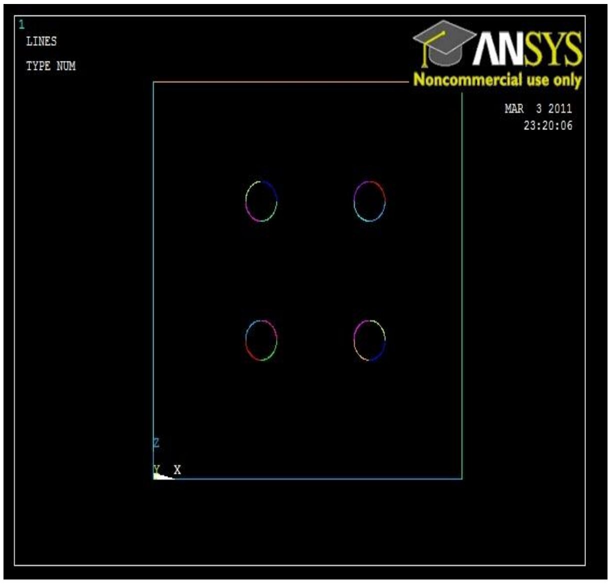
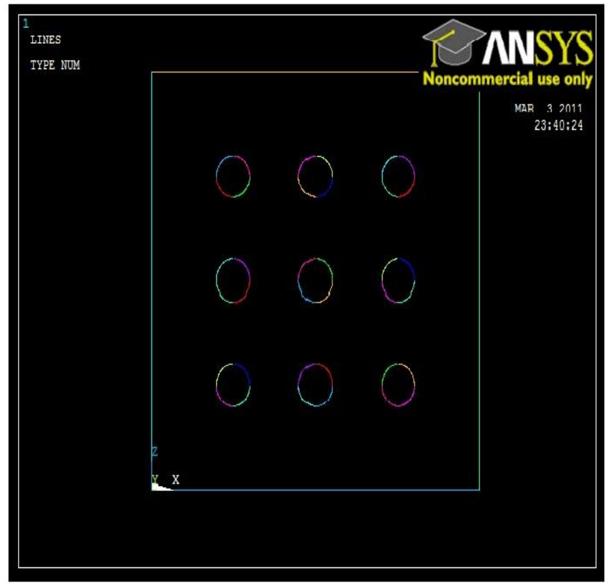


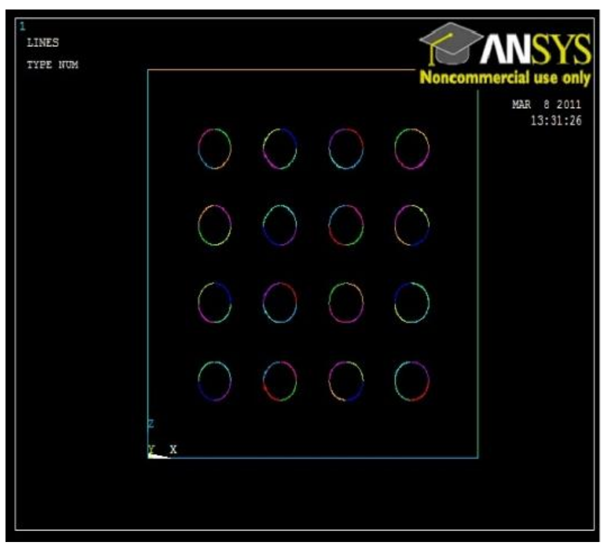
Fig.4. 1 Boundary conditions



(a)

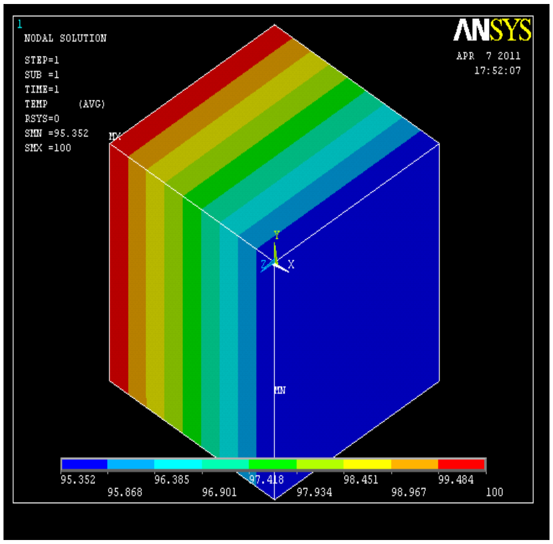


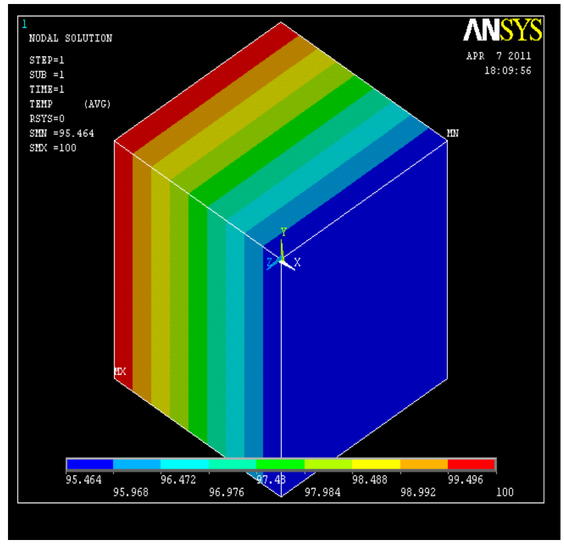
(b)



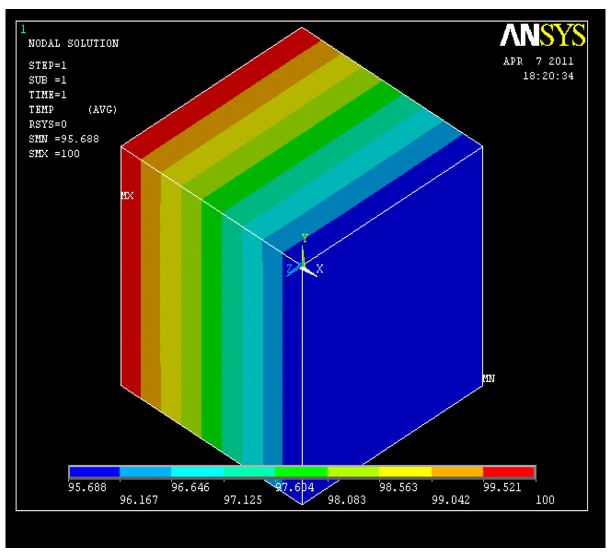
(c)

Fig. 4.2 Typical 3-D spheres-in-cube models with particle concentration of titania(TiO_2) powder (a) 0.4 vol% (b) 1.4 vol% and (c) 3.34 respectively.





(a) (b)



(c)

Fig. 4.3 Temperature profiles for Titania(TiO_2) filled epoxy composites with particle concentration of (a) 0.4 vol% (b) 1.4 vol% and (c) 3.34 vol% respectively

Sample	TiO_2 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	0.418	0.367	0.366
3	1.413	0.377	0.372
4	3.34	0.399	0.395

Table 4.2 Thermal conductivity for composites obtained from FEM and Experiment

Composite Sample	TiO_2 Content (Vol. %)	Percentage errors associated with FEM results w.r.t. the experimental value (%)
1	0.4188	0.273
2	1.413	1.344
3	3.34	1.012

Table 4.3 Percentage errors associated with the FEM simulated values with respect to the measured values (for TiO_2 filled epoxy composites)

GRAPH:

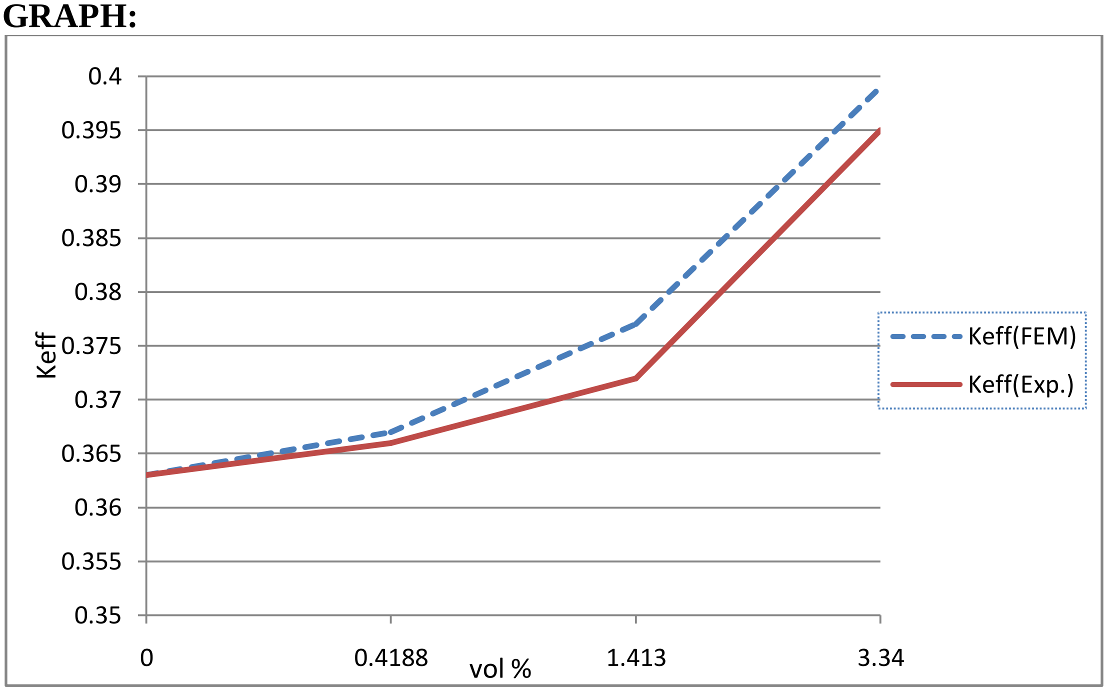


FIG. 4.4 Comparison between Experimental results and FEM Analysis

Thermal conductivities of epoxy composites filled with Titania(TiO_2)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, and 3.34 vol % are presented in Figures 4.3a - 4.3c 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 3.34 vol.%. Incorporation of TiO_2 results in enhancement of thermal conductivity of epoxy resin. With addition of 1.413 % and 3.34% of TiO_2 the thermal conductivity improves by about 3.17 and 9.022 % respectively when compared with 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 TiO_2 filled epoxy composites have led to the following specific conclusions:

- Successful fabrication of epoxy based composites filled with micro-sized TiO_2 by 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 3.34 vol.%.
- Incorporation of TiO_2 results in enhancement of thermal conductivity of epoxy resin. With addition of 3.34 vol. % of TiO_2, the thermal conductivity improves by about 9.002 % with respect to neat epoxy resin.
- These new class of TiO_2 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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References

1.Rajlakshmi Nayak, Dr. Alok satapathy(2010).A study on thermal conductivity of particulate reinforced epoxy composites.

2. S.W. Gregory, K.D. Freudenberg, P. Bhimaraj and L. S Schadler, A study on the friction and wear behavior of PTFE filled with alumina nanoparticles, J. Wear, 254 (2003) 573–580.

3. K. Jung-il, P.H. Kang and Y.C. Nho, Positive temperature coefficient behavior of polymer composites having a high melting temperature, J. Appl. Poly Sci., 92 (2004) 394–401.

4. S. Nikkeshi, M. Kudo and T. Masuko, Dynamic viscoelastic properties and thermal properties of powder-epoxy resin composites, J. Appl. Poly. Sci., 69 (1998) 2593-8.

5. K. Zhu and S. Schmauder, Prediction of the failure properties of short fiber reinforced composites with metal and polymer matrix, J. Comput. Mater. Sci., 28 (2003) 743–8.

6. M. Rusu, N. Sofian and D. Rusu, Mechanical and thermal properties of zinc powder filled high density polyethylene composites, J. Polymer Testing, 20 (2001) 409–17.

7. I. H. Tavman, Thermal and mechanical properties of copper powder filled poly (ethylene) composites, J. Powder Tech., 91 (1997) 63–7.

8. R.N. Rothon, Mineral fillers in thermoplastics: filler manufacture, J. Adhesion, 64 (1997) 87–109.

9. R.N. Rothon, Mineral fillers in thermoplastics: filler manufacture and characterization, J. Adv. Polym. Sci., 139 (1999) 67–107.

10. L.E. Nielsen and R.F. Landel, Mechanical properties of polymers and composites. second ed., Marcel Deckker, New York, 1994, pp.377–459.

11. S.T. Peters, Handbook of composites, second ed., Chapman and Hall, London, 1998, pp. 242–243.

12. R.J. Young and P.W.R. Beaumont, Failure of brittle polymers by slow crack growth Part 3 Effect of composition upon the fracture of silica particle-filled epoxy resin composites, J. Mater. Sci., 12(4) (1977) 684–92.

13. A.J. Kinoch, D.L. Maxwell and R.J. Young, The fracture of hybrid particulate composites, J. Mater. Sci., 20 (1985) 4169–84.

14. R. Young, D.L. Maxwell and A.J. Kinloch, The deformation of hybrid particulate composites. J. Mater. Sci., 21 (1986) 380–388.

15. S.W. Koh, J.K. Kim and Y.W. Mai, Fracture toughness and failure mechanisms in silica-filled epoxy resin composites: effects of temperature and loading rate, J. Polymer, 34(16) (1993) 3446–3455.

16. W.J. Cantwell and A.C. Moloney, Fractography and failure mechanisms of polymers and composites, Elesvier, Amsterdam (1994) 233.

17. M. Imanaka, Y. Takeuchi, Y. Nakamura, A. Nishimura and T. Lida, Fracture toughness of spherical silica-filled epoxy adhesives. Int. J. Adhesin Adhes., 21 (2001) 389–396.

18. H. Wang, Y. Bai, S. Lui, J. Wu and C.P. Wong, Combined effects of silica filler and its interface in epoxy resin, J. Acta. Mater., 50 (2002) 4369–4377.

19. I.Yamamoto, T. Higashihara and T. Kobayashi, Effect of silica-particle characteristics on impact/usual fatigue properties and evaluation of mechanical characteristics of silica-particle epoxy resins, Int. J. JSME, 46 (2) (2003) 145– 153.

20. Y. Nakamura, M. Yamaguchi, A. Kitayama, M. Okubo and T. Matsumoto, Effect of particle size on fracture toughness of epoxy resin filled with angularshaped silica, J. Polymer, 32(12) (1991) 2221–2229.

21. Y. Nakamura, M. Yamaguchi, M. Okubo and T. Matsumoto, Effect of particle size on impact properties of epoxy resin filled with angular shaped silica particles, J. Polymer, 32(16) (1991) 2976–2979.

22. Y. Nakamura, M. Yamaguchi, M. Okubo and T. Matsumoto, Effects of particle size on mechanical and impact properties of epoxy resin filled with spherical silica, J. Appl. Polym. Sci., 45 (1992) 1281–1289.

23. D. Hansen and C. Ho, Thermal Conductivity of High Polymers, J. of Poly. Sci. Part A, 3(2) (1965) 659–670.

24. S. Peng and R. Landel, Induced Anisotropy of Thermal Conductivity of Polymer Solids under Large Strains, J. Appl. Poly. Sci., 19(1) (1975) 49–68.

25. C.L. Choy, and K. Young, Thermal Conductivity of Semicrystalline Polymers– A Model, J. Polymer, 18(8) (1977) 769–776.

26. I. Tavman, Thermal Anisotropy of Polymers as a Function of their Molecular Orientation, Experimental Heat Transfer, Fluid Mechanics, and Thermodynamics, Elsevier (1991) 1562–1568,

27. R.C., Progelhof, J.L. Throne and R.R. Ruetsch, Methods of Predicting the Thermal Conductivity of Composite Systems, J. Polymer Engineering and Science, 16(9) (1976) 615–625.

28. N.S. Saxena, N.S. Pradeep Saxena, P. Pradeep, G., Mathew, S. Thomas, M. Gustafsson and S.E. Gustafsson, Thermal Conductivity of Styrene Butadiene Rubber Compounds with Natural Rubber Prophylactics Waste as Filler, J. European Polymer, 35(9) (1999) 1687–1693.

29. H. Ishida and S. Rimdusit, Very High Thermal Conductivity Obtained by Boron Nitride-filled Polybenzoxazine, Thermochimica Acta, 32(1–2) (1998) 177–186.

30. I. Tavman, Thermal and Mechanical Properties of Copper Powder Filled Poly (ethylene) Composites, J. Powder Tech., 91(1) (1997) 63–67.

31. A. Bjorneklett, L. Halbo, and H. Kristiansen, Thermal Conductivity of Epoxy Adhesives Filled with Silver Particles, Int. J. of Adhesion and Adhesives, 12(2) (1992) 99–104.

32. Y. Agari, A. Ueda, M. Tanaka and S. Nagai, Thermal Conductivity of a Polymer Filled with Particles in the Wide Range from Low to Super-high Volume Content, J. of Appl. Poly. Sci., 40(5–6) (1990) 929–941.

33. D. Kumlutas, I.H. Tavman, and M.T. Coban, Thermal Conductivity of Particle Filled Polyethylene Composite Materials, J. Composites Sci. and Tech., 63(1) (2003) 113–117.

34. D. Veyret, S. Cioulachtjian, L. Tadrist, and J. Pantaloni, Effective Thermal Conductivity of a Composite Material: A Numerical Approach, Transactions of the ASME- Journal of Heat Transfer, 115 (1993) 866–871.

35. J.T. Mottram and R. Taylor, Thermal Conductivity of Fibre/Phenolic Resin Composites. Part II: Numerical Evaluation, J. Composites Science and Technology, 29(3) (1987) 211–232.

36. O.O. Onyejekwe, Heat Conduction in Composite Media: A Boundary IntegralApproach, J. Computers & Chemical Engineering, 26(11) (2002) 1621– 1632.

37. I.H. Tavman, Thermal and Mechanical Properties of Aluminum Powder filled High-density Polyethylene Composites, Journal of App. Poly. Sci., 62(12) (1996) 2161–2167.

38. N.M. Sofian, M. Rusu, R. Neagu and E. Neagu, Metal Powder-filled Polyethylene Composites. V. Thermal Properties, J. of Thermoplastic Composite Materials, 14(1) (2001) 20–33.

39. H.S. Tekce, D. Kumlutas and I.H. Tavman, Determination of the Thermal Properties of Polyamide-6 (Nylon-6)/Copper Composite by Hot Disk Method,In: Proceedings of the 10th Denizli Material Symposium, (2004) 296–304.

40. R.C. Progelhof, J.L. Throne and R.R. Ruetsch, Methods of Predicting the Thermal Conductivity of Composite Systems, J. Polymer Engineering and Science, 16(9) (1976) 615–625.

41. P. Procter, J. Solc, Improved thermal conductivity in microelectronic encapsulants. IEEE Trans on Hybrids Manuf Technol, 14(4) (1991) 708–13.

42. Y. Nagai, G.C. Lai, Thermal conductivity of epoxy resin filled with particulate aluminum nitride powder, J. Ceram Soc Jpn, 105(3) (1997) 197–200.

43. A. Griesinger, W. Hurler and M. Pietralla, A Photothermal Method with Step Heating for Measuring the Thermal Diffusivity of Anisotropic Solids, Int. J. of Heat and Mass Transfer, 40(13) (1997) 3049–3058.

44. I. Tavman, Thermal Anisotropy of Polymers as a Function of their Molecular Orientation, Experimental Heat Transfer, Fluid Mechanics, and Thermodynamics, Elsevier (1991) 1562–1568.

45. N.M. Sofian, M. Rusu, R. Neagu and E. Neagu, Metal Powder-filled Polyethylene Composites. V. Thermal Properties, J. Thermoplastic Composite Materials, 14(1) (2001) 20–33.

46. Y.P. Mamunya, V.V. Davydenko, P. Pissis and E.V. Lebedev, Electrical and Thermal Conductivity of Polymers Filled with Metal Powders, J. European Polymer, 38(9) (2002) 1887–1897.

47. B. Weidenfeller, M. Ho¨ fer and F.R. Schilling, Thermal Conductivity, Thermal Diffusivity, and Specific Heat Capacity of Particle Filled Polypropylene, J. Composites Part A: Applied Science and Manufacturing, 35(4) (2004) 423–429.

48. H.S. Tekce, D. Kumlutas, and I.H. Tavman, Determination of the Thermal Properties of Polyamide-6 (Nylon-6)/Copper Composite by Hot Disk Method, In: Proceedings of the 10th Denizli Material Symposium, Denizli, (2004) 296– 304.

49. D. Kumlutas, and I.H. Tavman, A Numerical and Experimental Study on Thermal Conductivity of Particle Filled Polymer Composites, J. of Thermoplastic Composite Materials, 19 (2006) 441.

50. Patnaik Amar, Md. Abdulla, Satapathy Alok, B. Sandhyarani and K. S. Bhabani, A study on a possible correlation between thermal conductivity and wear resistance of particulate filled polymer composites, J. Materials and Design ,(In press) (2009).

51. H.J. Ott, Thermal Conductivity of Composite Materials, J. Plastic and Rubber Processing and Application, 1(1) (1981) 9–24.

52. Tsao T.N.G., Thermal Conductivity of Two Phase Materials, J. Industrial and Engineering Chemistry, 53(5) (1961) 395–397.

53. Cheng S.C. and Vachon R.I., The Prediction of the Thermal Conductivity of Two and Three Phase Solid Heterogeneous Mixtures, Int. J. of Heat Mass Transfer, 12(3) (1969) 249–264.

54. Y. Agari and T. Uno, Estimation on Thermal Conductivities of Filled Polymers, J. of App. Poly. Sci., 32(7) (1986) 5705–5712.

55. Y. Agari, A. Ueda and S. Nagai, Thermal Conductivity of Polyethylene Filled with Disoriented Short-cut Carbon Fibers, J. of Applied Polymer Composite Science, 43(6) (1991) 1117–1124.

56. J. Maxwell, Electricity and Magnetism, Oxford, Clarendon, 1873.

57. Agarwal B D, Broutman L J. Analysis and performance of fiber composites: Second Edition. John Wiley and Sons, Inc.; 1990.

58. M. J. Turner, R. W. Clough, H. C. Martin and L. J. Topp, Stiffness and Deflection Analysis of Complex Structures, J. of the Aeronautical Sciences, 23 (1956) 805-823.

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