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Thermal conductivity of particulate filled polymer composites

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ABSTRACT

Thermal conductivity of a polymer composite is a function of resin type, fiber type and filler content, direction of heat flow, and service temperature. Recognizing the thermal responses in Fiber Reinforced Polymer (FRP) composite play a critical role in their performance, accurate thermal measurements of FRP are essential. Thermal conductivity measurements are performed up to a filler concentration of 4wt% and 8 wt%. The objective of the research is to attain better understanding of global and local thermal behavior of a composite structural system through fundamental understanding of thermal conductivity properties. The literature reveals that a major focus has been placed on measurements of thermal conductivity in through-the-thickness direction of a FRP composite. Therefore, current research focus has been on characterization of thermal conductivity in the planar direction (along the direction of fiber and transverse direction of fiber) of a composite laminate. The characterization has been carried out using 'Guarded heat flow meter method' in accordance with ASTM E 1530. The results obtained from this study are useful for the prediction of thermal conductivity of pine bark reinforced epoxy resin composites with or with out filler content. The results can be used to provide guidance on filler selection for compounding for outdoor insulation applications. © 2009 Trade Science Inc. - INDIA

KEYWORDS

Polymer composite;
Thermal conductivity;
Particulate.

INTRODUCTION

Fiber Reinforced Polymer (FRP) composites is defined as a polymer (plastic) matrix, either thermoset or thermoplastic, that is reinforced (combined) with a fiber or other reinforcing material with a sufficient aspect ratio (length to thickness) to provide a discernable reinforcing function in one or more directions. FRP composites are different from traditional construction materials such as steel or aluminum. FRP composites are anisotropic (properties only apparent in the direction of the applied load) whereas steel or aluminum is isotropic (uniform properties in all directions, independent of applied load). Therefore, FRP composite properties are directional, meaning that the best mechanical properties are in the direction of the fiber placement. Composites are similar to reinforced concrete where the rebar is embedded in an isotropic matrix called concrete.

Composites are engineered materials made out of two or more components. Most of the composites can be tailored to obtain properties better than individual constituents. A polymer composite reinforced with fiber is called FRP composite. Considering a composite, which involves two or more macro constituent phases, the matrix is referred to as continuous phase and the fibers are called the reinforcing phase[1]. The fibers increase the strength and stiffness, increase thermal and fatigue properties, provide better dimensional stability and electrical resistivity. Whereas the primary function of the resin is to transfer load, to hold the fibers, protect fibers from environment and mechanical abrasion. Matrix also carries transverse loads and inter-laminar shear stresses[2]. The advantages of composites include high strength to weight ratio, non-corrosiveness, less maintenance, high electrical resistance, wear resistance, electromagnetic transparency, appeasing appearance etc. Composites can be manufactured in different ways depending upon matrix, reinforcement and application. Different manufacturing methods include Hand-lay up, Compression molding, Resin Transfer Molding (RTM), Pultrusion, Autoclave and Filament molding etc. Considering the mechanical and thermal properties of composites, they are anisotropic in nature (i.e. the properties vary with orientation), but most of the composites can be considered as orthotropic. This behavior of the material also causes the development of large values of thermal stresses due to temperature gradient across the section. Thermally conductive but electrically insulating, also for their cost-effectiveness and design flexibility, ceramic particle reinforced epoxy composites are largely used as electronic packaging and/or substrate materials[3,5]. During the last few decades, however, in the microelectronic systems, great effect has been contributed to improving higher integration density, faster performance, miniaturization of electronic devices and lower cost[6,7]. Therefore, the power density in the electronic devices is becoming larger and larger. That turns the researchers' focus to be placed on the thermal conductivity in order to get heat-dissipating composites. The prediction of thermal conductivity of composites comprises a significant portion of the heat transfer literature. Many reports concerning about the thermal conductivity of polymer composites, associated with various thermal conductive models or equations for predicting the thermal conductivity, have been published. They are either theoretically based or are empirical which means to include one or more experimentally determined (or empirical) parameters. A good overview has been given by Progelhof[8]. Procter and Solc[9] 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[10] found 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. Wong[4] found that Agari model predicts better than Maxwell model for thermal conductivity of SiO2/epoxy composite at high percentage of filler.

However, polymer composites are always not treated as different despite the volume fraction of the filler are not the same when their thermally conductivity are treated employing theoretical models mentioned above. At different volume fraction, the "filler" in the composite can be regarded to be either phase, for example: the dilute system when the polymer phase is regarded to be the matrix or the heavily loaded system when the filler can be considered to be the matrix. At the medial concentration, the fillers start to contact which is called the percolation phenomenon[11,12], then both phase can be treated as the matrix.

The objective of this present work is to understand the thermal behavior of FRP composites by developing a database of thermal conductivity for particulate filled FRP composite materials. Different composite samples are to be distinguished based upon the following parameters: different resin, effect of heat flow direction, fiber weight fraction, effect of different filler materials and different manufacturing methods. At the end, the experimental results are compared with exist theoretical model for validation purposes.

MATERIALS

Matrix material

The primary functions of the resin are to transfer stress between the reinforcing fibers, act as a glue to hold the fibers together, and protect the fibers from mechanical and environmental damage. Resins are divided into two major groups known as thermoset and thermoplastic. Thermoplastic resins become soft when heated, and may be shaped or molded while in a heated semi-fluid state and become rigid when cooled. Thermoset resins, on the other hand, are usually liquids or low melting point solids in their initial form.

Polyester resin

Unsaturated polyester resins (supplied by Ciba-Giegy Ltd. India) to prepare the composites. 2% cobalt nephthalate (as accelerator) is mixed thoroughly in isophthalic polyester resin and then 2% methyl-ethyl-ketone-peroxide (MEKP) as hardener is mixed in the resin prior to reinforcement. Polyester resin has modulus of 72.5 GPa and possess density of 1.35 gm/cc respectively.

Epoxy resin

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 corresponding hardener (HY951) 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.

Vinyl ester resin

Vinyl ester resins (EPN-1138, Ciba-Geigy Ltd., India) are addition products of various epoxide resins and unsaturated monocarboxylic acids, most commonly methacrylic acid[13]. They have terminal reactive double bonds derived from the carboxylic acid used. These reactive groups can form a cross linked network with or without the addition of a co-monomer. In many industrial products, vinyl ester resins are comprised of 40-50 wt% styrene. It is common to dilute the vinyl ester oligomers with a low molecular weight co-monomer, such as styrene, vinyl-toluene or methyl methacrylate, to reduce the room temperature viscosity of the mixture and yield a solution with a typical viscosity in the range of 200 to 2000 cps. The reaction to form the vinyl ester oligomers is usually catalyzed by tertiary amines, phosphines and alkali salts. Previous research shows that triphenyl phosphine is a more effective catalyst for this reaction as compared to other catalysts[14]. Typical reaction conditions are 120°C for 4-5 hours, and hydroquinone is commonly used as the initiator. The conversion of the reaction is 90-95 %.

Fibers material

Fiber is the reinforcing phase of a composite material. Thermal conductivity of a composite depends upon the thermal conductive nature of the fiber and matrix. Commonly used fibers for composites include Glass, Carbon, and Aramid etc. Glass fibers are commonly used for engineering composites. Their uses include the manufacturing of automotive parts, pipes, structural members etc. Glass fiber is available economically in abundance with good mechanical properties; thus widely used in composite structures[2]. Basing upon different applications glass fiber (silica-oxygen network) is classified into E glass, C glass, and S glass fibers[2]. E glass is used as an insulator and mostly used in electrical industry, hence got the name "E" before the word glass. E-glass also has good mechanical properties in addition to low cost and ease of usability. The letter "S" in S-glass stands for structural applications. S-glass got different chemical formulation and it has higher strength to weight ratio, and higher elongation strain percentage. S glass is a bit expensive and mostly used in structural application, and C-glass fibers are advantageous in resisting chemical corrosion. Glass fibers are available in different forms like continuous, chopped and woven fabrics.

Carbon fibers are manufactured using precursor materials like rayon, petroleum or coal tar pitches and polyacrylonitrile (PAN)[15]. The conversion of pitch or PAN precursor to carbon fibers involves manufacturing steps like fiber formation by spinning, stabilization to thermoset the fiber, carbonization, graphitization, surface treatment and sizing[1]. During graphitization stage at higher temperatures the crystallites are properly ordered and oriented along the axis direction of the fiber. In PAN based carbon fibers, during the graphitization stage the linear structure of carbon atoms transforms into a planar structure called as basal planes and are oriented or stacked along the axis of the fiber. These basal planes are closely packed and are responsible for the high modulus and higher electrical and thermal conductivities along the axis of the fiber.

Natural fibers are becoming potential alternatives for glass fiber reinforced composites in many applications[16]. Usage of natural fibers like hemp, jute, flax, cotton etc. instead of synthetic fibers leads to increase in specific properties like impact strength, crash behavior, sound absorption, thermal insulation and reduction in component's weight, pollutants, and greenhouse gas emissions making the composite more environmental friendly. Natural fiber composites have many applications including automobile parts like door trim panels, headliners or back panels. Natural fiber is filled with cellulose material, which acts as an insulator, thus a natural fiber composite shows much lesser thermal conductivity when compared to a glass fiber reinforced polymer (GFRP) composite.

Particulate materials

The primary advantage of using filler material in composites is to reduce the overall cost of the composite. In addition to reduction of cost, filler materials also serve as major ingredient, which improves the performance of the composite by enhancing crack resistance, reducing shrinkage, influencing mechanical strength, improving fire resistance etc. Major filler materials used in composite manufacturing are calcium carbonate, kaolin (clay), Aluminum-trihydrate etc.

Two conventional fillers: aluminium and copper powder and one industrial waste: cement by-pass dust (CBPD) are chosen to be used as particulate fillers in pre-determined proportions in various composites prepared for this investigation. While, aluminium and copper have conventionally been used in many composite applications, the waste cement by-pass dust (CBPD) can be considered as non-conventional materials for use as fillers in polymer composites.

Cement by-pass dust (CBPD) is a by-product of cement manufacturing. It is a fine powdery material similar in appearance to Portland cement. It is generated during the calcining process in the kiln. As the raw materials are heated in the kiln, dust particles are produced and then carried out with the exhaust gases at the upper end of the kiln. These gases are cooled and the accompanying dust particles are captured by efficient dust collection systems. Lime (CaO) constitutes about 40% of CBPD composition. Other compounds include SiO2, Al2O3, Fe2O3, K2O, Na2O, Cl-, etc.

MATERIALS AND SAMPLE PREPARATION

Test specimens are prepared using 0 %, 4 %, and 8 % of filler by weight percentages are taken in this work to determine conductivity properties and specific wear rate of the composites in three different directions (i.e. longitudinal, transverse, and through-the-thickness). The properties are function of composition parameters and process parameters. Herein, the thermal conductivity measurement in the transverse direction means that a sample is prepared in such a way that heat flow direction is transverse to primary fiber orientation of the sample; similarly, the thermal conductivity measurement in the longitudinal direction means heat flowing along the fiber direction; and the thermal conductivity measurement in the thickness direction means heat flowing through the thickness direction. The thermal conductivity of an anisotropic composite material depends on the resin nature, fiber type and architecture, fiber weight fraction, manufacturing technique, direction of heat flow and operating temperature, leading to a high degree of complexity.

Thermal conductivity test matrix

The lists of thermal conductivity test samples direction are given in TABLE 1. The test matrix has included various material parameters and process parameters, leading to a spectrum of composite samples for their thermal conductivities along 00 and 900 fiber orientations and through the thickness direction. It should be noted that sample preparation is the most challenging part of the entire process. Many tools are used specially for sample preparation in addition, attention to environmental and health safety issues are also needed.

The test samples are prepared in the form of 10cm rectangular plate using compression molding and hand lay-up methods. Ideally all samples for three-dimensional measurements should be cut from same block by diamond cutter. Technically this poses a great difficulty in manufacturing such a block in terms of proper alignment of fabric and wet out.

Typically, FRP laminate has a thickness of 3-8 mm. A 10cm rectangular plate is cut from this type of laminate to measure its thermal conductivity with heat flowing through the thickness direction. Both the above dis-

TABLE 1: List of thermal conductivity test sample directions

Effect of different materials and manufacturing methods					
Compression molding /Hand lay-up techniques					
Glass fiber / Carbon fiber reinforced composites			Pine bark reinforced composites		
L	T	Th	L	T	Th

L: Longitudinal direction, T: Transverse directions and Th: Through thickness

cussed approaches are used to prepare samples in the present study. As a result, we have successfully prepared a couple of samples that allow for thermal conductivity measurements in transverse, longitudinal, and through the thickness directions.

Composite preparation

In addition to these, E-glass fiber / carbon fiber are reinforced with respective resin also added particulate filler materials to prepare samples with different wt%. All these filler materials are thoroughly dispersed in the resin by mechanical stirring. The resins used for this study were vinyl ester, polyester and epoxy. Vinyl ester resin is obtained from by Ciba-Geigy of India, Ltd. To prepare vinyl ester neat resin and neat epoxy resin samples, 2% Methyl Ethyl Ketone Peroxide (MEKP) is used as a catalyst. For polyester sample, the polyester resin is mixed with 2% MEKP (catalyst) and 4% Cobalt Napthanate (promoter). The promoter is mixed in the resin thoroughly before adding the initiator. The reaction between initiator and promoter caused the resin to become gel. Since the reaction between initiator and promoter is very fast. Care should be taken while preparing the samples in order to avoid voids. The carbon fiber /vinyl ester composite sheet was hand-laid up and then compressed. A carbon/vinyl ester composite block is prepared to generate all test specimens in three directions, i.e. through the thickness, longitudinal and transverse directions. Similarly, other composites are prepared both hand lay-up and compression molding techniques.

Natural fiber /vinyl ester composites

In this study, a natural fiber composite material is tested for its thermal conductivity in a limited manner because of its growing importance[16]. This natural fiber composite plate is made of four layers of natural fiber mat wet out with resin by hand-lay up process and cut into different dimensions for thermal conductivity measurement in all the three directions.

Factors affecting thermal conductivity of composite materials

Thermal conductivity of composites is anisotropic in nature. The knowledge of thermal conductivity of composites is needed for accurate design. Data about thermal conductivity of resin facilitates to reduce stresses related to shrinkage of composites during cure and mismatch in thermal expansion coefficients. Before conducting experiments to determine thermal conductivity of various composites, knowledge about effect of different parameters influencing thermal conductivity is essential.

Manufacturing methods

As the thermal conductivity of a polymer composite is based upon the conductivity of fiber and resin, being a polymer, resins are usually insulating and the conductivity is dominated by fiber material. The compactness of fibers per unit area influences the conductivity of the composite. Fiber packing in a composite depends on the method of manufacturing. The various composites manufacturing techniques are Hand lay-up, Compression molding, Resin Transfer Molding, Pultrusion etc[2].

Hand lay-up techniques

Hand lay-up technique is the oldest and simplest method of manufacturing composites. The tools required for the process are a mold to accommodate dry manufacturing according to the desired shape and a roller to facilitate uniform distribution of resin. Virtually any sized composites can be manufactured using this method. This method is the cheapest method of manufacturing but it has some disadvantages such as long curing time, low production rate, and further the quality of the composite depends on the skill of the worker.

Compression molding process

This method is commonly used to manufacture sheet molding or bulk molding. Compression molding machine consists of a male and female dies or platens to form the mold. The reinforcement combined with resin is placed in the mold and a hydraulic press is used to apply high pressure by closing male and female halves of the mold. After the material is cured, the pressure is released and the part is removed from the mold. Exterior body panels for structural members such as automobile bumpers are widely manufactured using this method.

Experimental approach for thermal conductivity measurements

There are various test methods available for thermal conductivity measurement and each method depends mainly on configuration of a material and job requirement. Some principal experimental methods for determinations of thermal conductivity are discussed below:

Guarded heat flow meter

This method is based on two dimensional steady state techniques and is used to measure and compare thermal properties of materials under controlled conditions and their ability to maintain required thermal conductance levels[16]. The specimen and a heat flux transducer (HFT) are sandwiched between two flat plates controlled at different temperatures, to produce a heat flow through the stack. A cylindrical guard surrounds the test stack and is maintained at a uniform mean temperature of the two plates, in order to minimize the lateral leak of heat. At steady state, the difference in temperature between the surfaces contacting the specimen is measured with temperature sensors embedded in the surfaces, together with the electrical output of the HFT. The output voltage is proportional to the heat flow through the specimen, HFT and the interfaces between the specimen and the apparatus. The coefficient of thermal conductivity can be obtained by prior calibration of system with the specimens of know thermal conductivity. At equilibrium, the thermal conductivity of material can be given as:

$$K = \Lambda x / R_s \qquad (1)$$

$$R_s = (N(\Delta T)/Q) - R_0 \qquad (2)$$

where K is the thermal conductivity (W/ m-K), R is the resistance of unknown specimen (m^2.K/ W), N is the HFT calibration constant, ΔT is the temperature difference between one surface of the specimen and the other surface (K), Δx is the specimen thickness (m), and R is the contact thermal resistance.

Experimental setup

A guarded heat flow meter method has been developed for thermal conductivity measurements. This is achieved by using a thermal conductivity testing system Unitherm model 2022 from ANTER Corp., Pittsburgh, PA. The experimental set up is shown in Figure 1.

This unit is supplied with a mid range flux module covering a thermal resistance range from 0.002 to 0.02 m^2 K/W and is able to measure the thermal conductivity of materials in the range of 0.1 to 40 W/m-K in terms of test standard ASTM E 1530. The materials that can be tested include metals, ceramics, polymers, composites, glass etc. The test samples need to be prepared in a form of two-inch diameter circular discs with their thickness depending on the materials' thermal conductivity. The thermal conductivity machine was supplied with three sets of calibration samples span the R_s range from 0.0005 to 0.05 m^2K/W. These samples were tested for conductivity values and compared with the given values by the manufacturer for calibration purpose.

Figure 1: Experimental set up of unitherm 2022

This equipment requires compressed air to raise and lower the upper stack assembly. It also needs either city water or a chiller to cool the heat sink, giving an operation temperature range from 20^0C to 300^0C. ANTER model Unitherm 2022 is a computerized system. The computer automatically controls the equipment for testing and data processing through a latest version of electronics and operating software once a test program is designed and initiated.

A schematic picture shown in Figure 2 represents the system functioning in detail. The assembly is a stack of parts with different functionalities. The heater on top and bottom helps to maintain steady state heat transfer through the sample, two polished surfaces on top and bottom the sample transfer heat from top and bottom of heaters with reduced thermal resistance through surface. A reference calorimeter is placed under the lower plate, which acts as a heat flux transducer. The heat

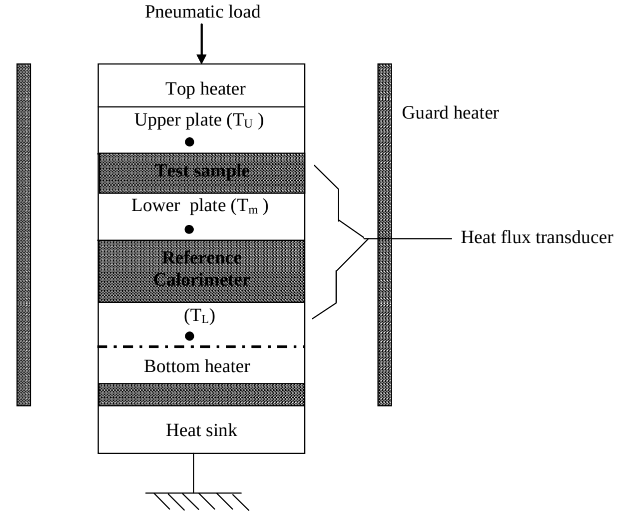

Figure 2: Schematic model showing the system arrangement in unitherm 2022

sink at the bottom avoids excessive temperature from the system. The sample is compressed in between the polished surfaces, each controlled at different temperatures, using pneumatic load. The pressure is maintained at 10 psi using pressurized air supply. As an option, coolant water is circulated through heat sink. A circular low temperature heat insulation ring is wrapped around the lower stack of the assembly to restrain heat flow to outside atmosphere. The entire system is maintained in a thermally insulated glass chamber. Unitherm is completely automatic while testing as the apparatus is completely controlled by a computer. The test system is hooked to the computer by means of an USB cable. Considering the operation of the system, the sample to be tested is prepared into a flat surface on both sides and thermal compound is applied on the sample to reduce thermal resistance caused due to surface roughness. Then the prepared sample is placed in between two polished surfaces and a pneumatic pressure of 10 psi is applied on the top portion of the stack. The sample can be tested in the temperature range from 20^{0}C to 300^{0}C. For steady state heat transfer, the user can divide the testing into different zones, called as set point temperatures. In this thesis the testing was divided into three temperature zones i.e. 60, 80, 100 ^{0}C. For higher thermal conductivity materials, a difference of 50 degrees is recommended for the machine. At every set point temperature, the system checks for steady state heat flow through the sample and thermal conductivity is measured.

Operation principle of unitherm 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.

$$Q = KA\frac{T_1 - T_2}{x} \qquad (3)$$

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 equation 4

$$R = \frac{T_1 - T_2}{Q/A} \qquad (4)$$

Where, R is the resistance of the sample between hot and cold surfaces (m^2-K/W). From Equations 3 and 4 we can derive that

$$K = \frac{x}{R} \qquad (5)$$

TABLE 2: Thermal conductivity values of different resins

Sample	Thickness (mm)	Thermal conductivity (W/m-K)		
		Test-1 (60 ^{0}C)	Test-2 (80 ^{0}C)	Test-3 (100^{0}C)
Neat polyester resin	0.567	0.196	0.215	0.24
Neat Epoxy resin	0.743	0.221	0.237	0.263
Vinyl ester resin	0.687	0.185	0.19	0.213

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.

EXPERIMENTAL RESULTS AND DISCUSSION

Neat resin castings and resin castings with fillers

The thermal conductivity of a composite depends on many parameters including 1) Direction of fiber; 2) Specific gravity of fiber; 3) Moisture content; 4) Distribution of moisture; 5) Voids and 6) Resin type. The

TABLE 3: Thermal conductivity of epoxy resin with different particulate fillers

Sr. No.	Sample	Thickness (mm)	Thermal conductivity (W/m-K)		
			Test-1 (60^{0}C)	Test-2 (80^{0}C)	Test-3 (100^{0}C)
1	Neat epoxy resin	0.743	0.221	0.237	0.263
2	4 wt% of Aluminium filled epoxy composite	2.114	0.347	0.359	0.351
3	8 wt% of Aluminium filled epoxy composite	3.764	0.489	0.534	0.514
4	4 wt% of copper powder filled epoxy composite	2.734	0.393	0.412	0.422
5	8 wt% of copper powder filled epoxy composite	3.987	0.372	0.396	0.393
6	4 wt% of CBPD filled epoxy composite	2.152	0.239	0.197	0.190
7	8 wt% of CBPD filled epoxy composite	3.634	0.227	0.228	0.229

parameters of major influence on thermal conductivity are fiber weight percentage and conductivity properties of both resin and fiber. In this present study, thermal conductivities of different resins like vinyl ester, polyester and epoxy are determined and the results are in agreement with the literature [17]. The results are listed in TABLE 2 and graphically presented in Figure 3a.

Fillers like aluminium, copper powder and cement by-pass dust are directly mixed with the epoxy resin and the effect of addition of these fillers on thermal conductivity is determined. Different samples with 4 wt%, 8 wt% of aluminium, 4 wt%, 8 wt% of copper powder and similarly for CBPD fillers are mixed in the epoxy resin for testing and results are listed in TABLE 3 and graphically plotted in Figures 3b, 3c and 3d.

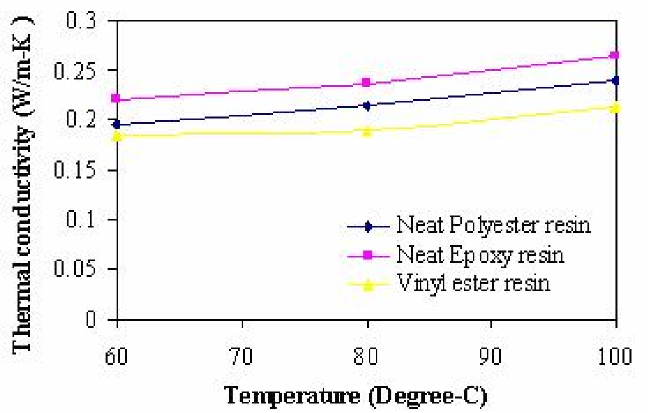

Figure 3a : Thermal conductivity variations with different resins

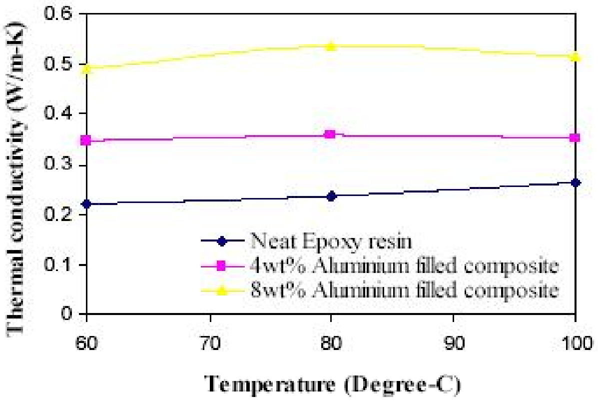

Figure 3b :Thermal conductivity variations with temperatures of aluminum filled epoxy composites.

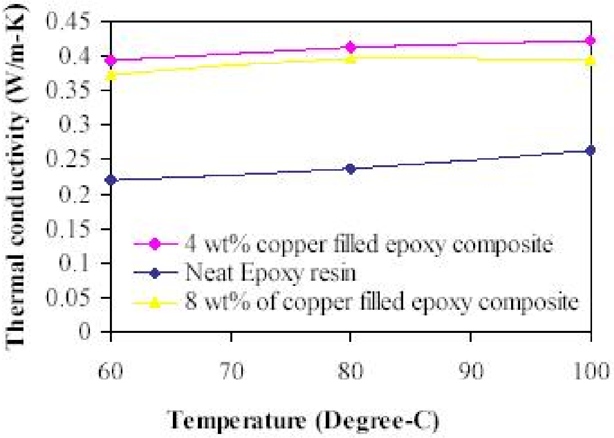

Figure 3c : Thermal conductivity variations with temperatures of copper filled epoxy composites.

The results show that the thermal conductivity values of vinyl ester, polyester, and epoxy have nearly the same values. As the vinyl ester sample has little air voids, it is showing lower thermal conductivity than the other two resins. Addition of aluminium particulate under three different percentage (0wt%, 4 wt% and 8 wt%) in the epoxy matrix composite significantly increases the thermal conductivity as shown in Figure 3b. For example, 4 wt% and 8 wt% respectively result in 64% and 45% increase in thermal conductivity over neat resin for temperatures ranging from 60-100^{0}C. Addition of 4 wt% and 8 wt% of copper powder in neat epoxy resin resulted in increase of conductivity values by 56% and 59% respectively (Figure 3c). However, for CBPD particle with epoxy resin composite, slight increase in thermal conductivity is noticed at the temperature segment of 60^{0}C when compared to the neat epoxy resin; but the thermal conductivity almost remains unchanged at the temperature range of 80^{0}C to 100^{0}C. This could have happened because addition of CBPD particle makes the sample brittle and hairline cracks were formed in the sample under heat and pressure. These cracks create air voids thus reducing the heat capacity and corresponding thermal conductivity. These results are tabu-

TABLE 4 : Thermal conductivity of E-glass fiber / epoxy resin compositestures of CBPD filled epoxy composites.

Sample	Manufacturing method	Thickness (mm)	Thermal conductivity (W/m-K) Test-1 (60 ^{0}C)	Thermal conductivity (W/m-K) Test-2 (80 ^{0}C)	Thermal conductivity (W/m-K) Test-3 (100^{0}C)
E-glass fiber + epoxy resin composite	Hand-lay-up technique	5.663	0.304	0.334	0.369
E-glass fiber + epoxy resin composite	Compression molding machine	5.225	0.372	0.389	0.412

TABLE 5 : Thermal conductivity of E-glass fiber / Epoxy resin composites

Heat flow direction	Thickness (mm)	Thermal conductivity (W/m-K) Test-1 (60 ^{0}C)	Thermal conductivity (W/m-K) Test-2 (80 ^{0}C)	Thermal conductivity (W/m-K) Test-3 (100^{0}C)
Transverse direction	3.228	0.279	0.297	0.334
Longitudinal direction	4.300	0.246	0.271	0.301
Through the thickness	5.663	0.320	0.340	0.357

lated in TABLE 3 and plotted in Figure 3d.

E-glass /epoxy resin composites

Thermal conductivity measurements of E-glass/epoxy samples are manufactured in two different methodologies i.e. by Hand-lay-up technique and Compression molding method are reported in TABLE 4. Thermal conductivity data for these samples are obtained by testing in through-the-thickness direction. For glass/epoxy composites, manufacturing method has little effect on thermal conductivity. Experimental results show that compression molding method samples have higher thermal conductivity than the other method. The potential cause for this variation in thermal conductivity might be due to air voids. Two potential reasons are suggested for formation of air voids: 1) stitching closely packs the fabric layers, leaving small air pockets between the layers, and applied resin might not seep through the air gaps fully, these air pockets hinder full conductivity; and 2) during composite fabrication by hand lay up technique the formation of holes can not be neglected and may even formation of a thin interfacial gap between the fiber and resin due to inadequate interfacial compatibility acting like air void and in turn reducing the thermal conductivity. These factors potentially change thermal conductivity by around ±0.05 W/ m-K. For all samples thermal conductivity increases slightly with test increased temperature.

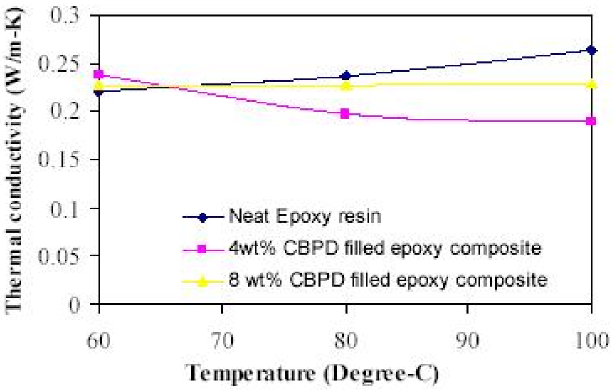

Figure 3d : Thermal conductivity variations with temperatures of CBPD filled epoxy composites.

E-glass/epoxy resin composite block

The thermal conductivity values of E-glass/ epoxy resin composite blocks in longitudinal, transverse and through-the-thickness directions are listed in TABLE 5. It is observed that the thermal conductivity in through-the-thickness direction is higher than that in the transverse direction, which in turn is higher than the conductivity in longitudinal direction. The conductivity value is supposed to be higher in longitudinal direction. However, in this case, bi-axial fabric might have led to this anomaly. Further more, the sample preparation also influences the results. After cutting the sample in 0 and 90 directions the fiber would be in the direction of heat flow and these fibers form a corrugated shape on the top and bottom surfaces. According to the sample requirements of the test facility, these surfaces are supposed to be smooth with a tolerance of ±0.005 inch, which is difficult to achieve with the available resources. However, the thermal conductivity value of the E-glass/epoxy resin block showed no significant change with respect to the direction of fiber. The fiber weight fraction for this block is determined as 49.8%. The low thermal conductivity of balanced biaxial E-glass fabric in 0 and 90 directions (i.e. in horizontal plane) resulted in heat conduction to be nearly uniform in both longitudinal and transverse directions.

TABLE 6 : Thermal conductivity of carbon fiber / epoxy resin composites

Heat flow direction	Thickness (mm)	Thermal conductivity (W/m-K)		
		Test-1 (60 ^{0}C)	Test-2 (80 ^{0}C)	Test-3 (100^{0}C)
Carbon fiber reinforced epoxy resin composite for 40 % fiber loading				
Through the thickness	3.772	0.245	0.262	0.283
Transverse direction	4.325	0.457	0.500	0.541
Longitudinal direction	5.176	0.787	0.965	1.231
Carbon fiber reinforced epoxy resin composite for 60 % fiber loading				
Through the thickness	4.165	0.312	0.320	0.337
Transverse direction	5.179	0.429	0.447	0.463
Longitudinal direction	5.664	1.367	1.941	2.227

Carbon fiber /epoxy resin composites

The thermal conductivity values of carbon fiber/ epoxy resin composite materials in longitudinal, transverse and through-the-thickness directions are listed in TABLE 6. The following observations are made from the test results of carbon fiber/epoxy resin composite material: 1) the thermal conductivities of carbon composites are the highest along the fiber direction, medium in the transverse direction and lowest in through-the- thickness direction; 2) carbon composites have significantly different thermal conductivity values in three directions – strongly anisotropic. This is distinctively different from E-glass composite materials for which thermal conductivities in three directions are close - almost isotropic; 3) thermal conductivity of carbon composites through the thickness direction is close to that of neat resin, indicating that thermal property of the resin plays a major role in this direction; 4) Carbon compos-

TABLE 7 : Thermal conductivity of natural fiber / Epoxy resin composites

Heat flow direction	Thickness (mm)	Thermal conductivity (W/m-K)		
		Test-1 (60 ^{0}C)	Test-2 (80 ^{0}C)	Test-3 (100^{0}C)
Natural fiber reinforced epoxy resin composite for 40 % fiber loading				
Neat epoxy resin	0.743	0.221	0.237	0.263
Through the thickness	3.442	0.123	0.128	0.136
Transverse direction	4.128	0.157	0.161	0.155
Longitudinal direction	4.176	0.175	0.190	0.208
Natural fiber reinforced epoxy resin composite for 60 % fiber loading				
Neat epoxy resin	0.743	0.221	0.237	0.263
Through the thickness	3.853	0.130	0.142	0.198
Transverse direction	4.469	0.177	0.185	0.189
Longitudinal direction	5.102	0.202	0.216	0.228

TABLE 8 : Theoretical models for transverse/ longitudinal effective thermal conductivity

Sl. no	Author	Model	Notes
Transverse effective thermal conductivity			
1.	Inverse rule of mixture	$\frac{1}{K_T} = \frac{V_f}{K_f} + \frac{V_m}{K_m}$	Derived from rule of mixtures to calculate for transverse thermal conductivity Does not consider voids, fabric overlap
Longitudinal effective thermal conductivity			
2.	Rule of mixture	$K_1 = K_f V_f + K_m V_m$	For longitudinal effective thermal conductivity
Thermal conductivity of the particulate filled pine bark polyester composites			
3.	Hamilton-Crosser formula	$K_c = K_M \frac{K_F + (n-1)K_M - (n-1)f(K_M - K_F)}{K_F + (n-1)K_M + f(K_M - K_F)}$	

ites result in lower thermal conductivity values in through-the-thickness direction than glass fiber composite. This observation is in accordance with the results published in the literature[18] and 5) as shown in TABLE 5, through-the-thickness thermal conductivity of carbon/ epoxy resin composite for 40wt% is consistent with that of carbon/ epoxy resin composite for 60wt%.

Carbon fiber is orthotropic in nature and the conductivity of the carbon fiber along the axis would be higher than in other directions because of the basal plane formation along the axis. These basal planes are closely packed in the order of angstrom units, which helps in transferring heat quickly and by virtue of its property carbon fibers contracts upon heat due to negative expansion coefficient further reducing the distance between the basal planes. Thus thermal conductivity increased linearly in longitudinal direction in the measured temperature range of 60-100^0 C. In transverse direction, conductivity would be much lower than that of in longitudinal direction due to lack of basal planes. Moreover, carbon sheets would be sized to have good compatibility with the resin, and this sizing causes an insulating layer and thus hinders heat flow in case of through the thickness direction. This glossy sizing over the carbon tow might be the reason for lower conductivity of carbon composite in through the thickness direction than that of a glass fiber composite. To test the effect of fiber weight fraction of a composite on thermal conductivity, another carbon/epoxy resin block was made with higher fiber compactness and tested. The results are also included in TABLE 6. The results are compared graphically in Figure 4.

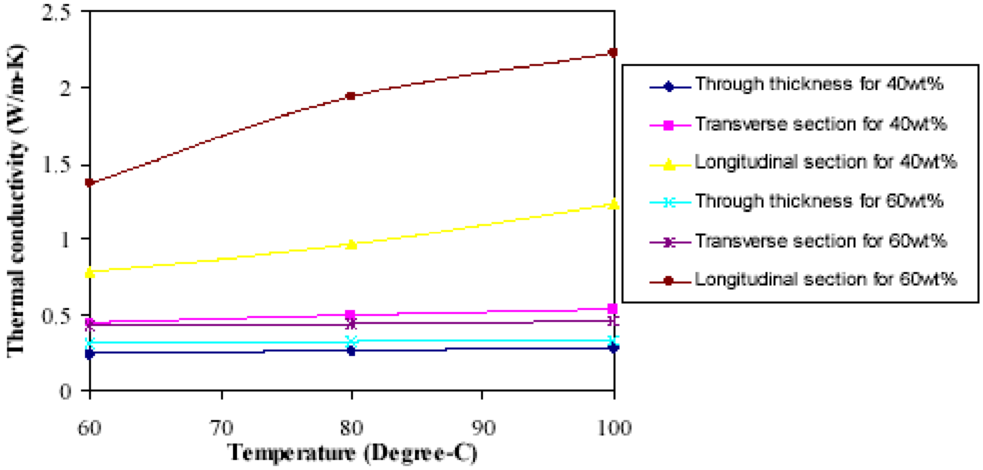

Figure 4 : Thermal conductivity variations for carbon fiber/ epoxy resin composites

Natural fiber /epoxy resin composites

The thermal conductivity of natural fiber reinforced epoxy composites have an extremely low thermal conductivity as shown in TABLE 7 and also shown in Figure 5, much lower than neat epoxy, demonstrating excellent thermal insulation characteristics.

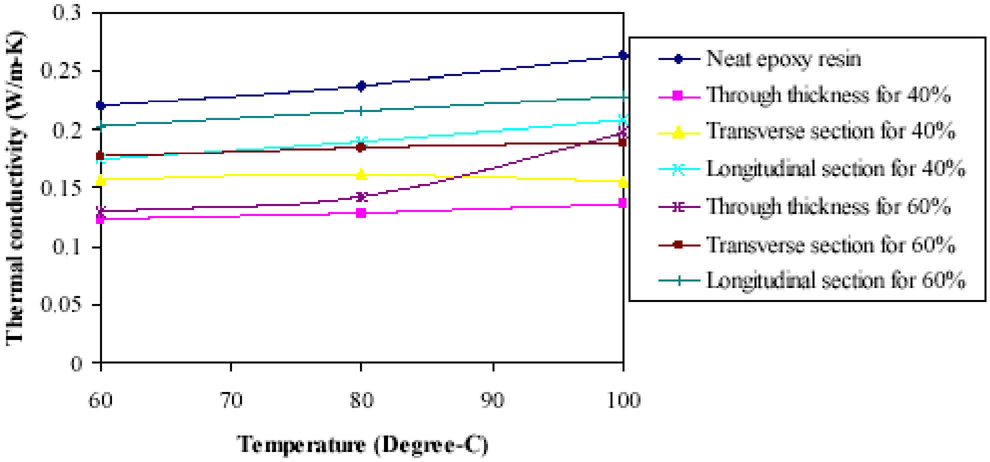

Figure 5 : Thermal conductivity variations for natural fiber/ epoxy resin composites.

TABLE 9 : Thermal conductivity of the samples using theoretical model

Composites	K_f (along the axis of fiber)	K_f (along transverse direction)	K_m	V_f (%)	V_m (%)
E-glass fiber / polyester resin	0.7	0.6	0.263	50	50
Carbon fiber / Polyester resin	8	0.54	0.263	50	50
Pine bark / Plyester resin	0.3	0.36	0.263	50	50
CBPD/Polyester resin	0.74	0.63	0.263	7	93
Pinebark/CBPD /Polyester	0.5	0.5	0.263	36.4	63.4
Pinebark/CBPD /Polyester	0.5	0.5	0.263	26.1	73.9

TABLE 10 : Comparison between experimental values and theoretical values

Method/ Model	Manufacturing Techniques	Pine bark/ Epoxy resin	Epoxy resin /CBPD	Pine bark/ CBPD/Epoxy resin
Experimental	Hand lay-up techniques	0.272	0.268	0.196
Inverse rule of mixture	Hand lay-up techniques	0.281	0.293	--
Hamilton-Crosser formula	Hand lay-up techniques	---	---	0.206
Error (%)		3.20%	10.23%	4.85%

VALIDATION OF MODEL PREDICTIONS

Analytical or numerical models help to predict the properties of a material without conducting any experiments. However, these models have to be extensively validated with experimental data before adopting them in practice on a large scale. Most of the literature on composite materials (anisotropic) dealt with mechanical properties and very few models were developed to predict thermal conductive properties in different directions. Most of the literature predicts through-the-thickness effective thermal conductivity, while thermal conductivity along the fiber direction in a composite can be predicted using rule of mixtures. Experimental results obtained for the Pine bark / epoxy resin samples are validated for through-the-thickness thermal conductivity and longitudinal thermal conductivity. Thermal conductivities through-the-thickness and longitudinal directions are verified using the theoretical models described in TABLE 8. In this study, thermal conductivity considered for validation purpose is the value obtained at a temperature of 100^0C. The details of thermal conductivity about the samples are given in TABLE 9 along with fiber and matrix volume fractions respectively.

In the present case thermal conductivity in transverse / longitudinal direction is calculated for pine bark reinforced with epoxy resin, CBPD reinforced with epoxy resin and combined all the three phases i.e pine bark reinforced with epoxy resin filled with cement by-pass dust taken both experimental and theoretical model for validation purposes. Comparisons between experimental and theoretical values are shown in TABLE 10. The obtained experimental results are in good agreement (~10%) with the theoretical models.

CONCLUSIONS

1. In case of thermal characterization of composite materials, few theoretical and limited experimental methods are available.
2. Most of the models do not consider for thermal resistance, interaction between fiber and matrix and filler content.
3. The thermal conductivity values of three different matrices Polyester, Epoxy resin and vinyl ester resin given as 0.196, 0.221 and 0.185 respectively thus showing nearly the same thermal conductivity values.
4. With the addition of 4 wt% and 8 wt% of aluminium additive in neat epoxy resin increased the conductivity by 64% and 45% respectively. Addition of 4 wt% and 8 wt% of copper powder in neat epoxy resin resulted in increase of conductivity values by 56% and 59% respectively. It is also observed that by the addition of industrial waste like cement by-pass dust with neat epoxy resin the thermal conductivity value for 4wt % of CBPD is increases around 92% and for 8wt% CBPD is around 97% as compared with neat epoxy.
5. In case of E-glass/Epoxy resin composite, the conductivity is increases from 0.221 to 0.320W/m-K in through-the-thickness direction than in transverse and longitudinal directions.
6. The thermal conductivity along longitudinal direction in case of Carbon/Epoxy resin composite is

almost twice the conductivity in transverse and three times greater than through-the-thickness direction. But in case of natural fiber reinforced epoxy composite there is no dramatic change in thermal conductivity values as compared to glass fiber/ carbon fiber reinforced epoxy composites.

7. Thermal conductivity of ceramic particles filled natural fiber reinforced epoxy composites are also discussed Hamilton-Crosser formula for validation purposes.
8. Some theoretical models available are of no good use for the validation of experimental results, as they require various properties of fibers and matrix, which are difficult to acquire for practical purpose.
9. Experimental results obtained from E-glass/Epoxy resin sample in longitudinal and through-the-thickness direction are in good agreement with the values obtained from theoretical models. In longitudinal direction, rules of mixture showed an upper bound value and in through-the-thickness direction inverse rule of mixture showed a lower bound experimental data.

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