Evaluation of Thermal Properties of E-Glass/ Epoxy Composites Filled By Different Filler Materials

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Abstract
This paper compares the values of the thermal and fire resistance properties of composites made using the hand layup technique. E-Glass fiber reinforced epoxy composites was fabricated by filling varying concentration of aluminum oxide (Al_2O_3), magnesium hydroxide($Mg(OH)_2$), silicon carbide (SiC), and hematite powder. The main aim of this work was to determine the thermal conductivity, thermal expansion coefficient, time to ignition and flame propagation rate of composites. Experimental results show that Al_2O_3 and Mg $(OH)_2$ filled composites exhibited low thermal conductivities. Composites filled by SiC particles exhibited low thermal expansion coefficient when compared with other filled composites. Fire test results indicated that increase the loading of Al_2O_3, Mg $(OH)_2$, and hematite powder increase the time to ignition and reduces the flame propagation rate of composites.

Key words: Composites, Fillers, Fire resistance, Properties, Thermal

1. Introduction
The primary field of application for fiber reinforced polymer composites is the aerospace industry. For several years, however, composite materials have been increasingly used for various other technical tasks, where it is beneficial to apply lightweight construction materials which have high strength and stiffness characteristics. The favorable specific properties of fiber reinforced polymer composites are based on the low density of the matrix resins used, and the high strength of the embedded fibers. Fabrication of fiber reinforced polymer composites relatively low cost. These composites are considered as replacements for metal materials. Polymer matrix reinforced by fiber is probably the most commonly used form of composites in structural application, such as air craft's, boats, automobiles[1] etc. For many materials applications, information is needed on their thermal properties [2]. In recent years there have been an increasing number of applications such as communication, satellites, high density electronics, and advanced aircraft requiring more effective and light weight thermal management materials [3]. The temperature fields in composites materials cannot be determined unless the thermal conductivity of the media is known and for any material a low thermal expansion is ideally required [2, 4]. Polymer composites are very flammable due to their chemical structure and they cannot satisfy some applications, which require high flame retardency. H. Dvir et al. [5] have studied the effect of additives such as pentabromobenzyleacryllate (PBBMA) and magnesium hydroxide on mechanical and flame retardant properties of polypropylene composites containing glass fibers. He concluded that addition of PBBMA and Mg (OH_2) has a positive effect on flame retardant properties and with minimal negative effect on mechanical properties and optimized the formulation of percentage of matrix and reinforcement. The best formulation obtained was with 38.50% matrix 22.31 % fibers and 15 % PBBMA and 17.99 % Mg (oH_2). It has good impact strength 90+.4 and modulus 6953mpa, Vo UL-94 rating and total flaming time 6sec. A. Shojaei et al. [7] have determined the thermal conductivity of rubber-based composites friction materials. Various thermally conductive fillers including copper, brass chips with two sizes, aluminum chips, aluminum oxide and talc have been selected and the effect of addition of these fillers on the thermal conductivity. It was found that the thermal conductivity of the materials increases continuously by increasing the content of the selected fillers. Maximum thermal conductivity is related to aluminum chips filled composites. Now a variety of inorganic fillers have been used to change the properties of composites. In this work E-glass/epoxy based composites filled with varying concentrations of aluminum oxide (Al_2O_3), magnesium hydroxide (Mg $(OH)_2$), silicon carbide (SiC), and hematite powder were prepared by hand layup technique. The objective of this work was to investigate the effect of fillers on thermal and fire resistance properties of E-glass fiber reinforced epoxy composites and comparison of results.

2. Materials and Fabrication
2.1. Materials
The composites were made from E-glass fiber and commercially available ARALDITE (L-12) along with hardener K-6. Al_2O_3, Mg $(OH)_2$, SiC and hematite powder was used as filler materials. Aluminum oxide particles is a ceramic powder commonly used filler, it is also used as an abrasive due to its hardness. Magnesium hydroxide is an inorganic compound and it is a white powder with specific gravity of 2.36, very slightly soluble in water; decomposing at 350^o C. Magnesium hydroxide is attracting attention because of its performance, price, low corrosiveness and low toxicity. Silicon carbide exhibits favorable mechanical and chemical properties at high temperatures for many applications. The benefits of using

SiC as reinforcement are improved stiffness, strength, thermal conductivity, wear resistance, fatigue resistance, reduced thermal expansion and dimensional stability. Hematite is an iron oxide with the same crystal structure as that of corundum (rubies and sapphires). Usually the color varies between a metallic grey and red. Hematite is one of the oldest stones mined in our history.

2.2 Fabrication of Composites

The E-glass /Epoxy based composites filled with varying concentrations (0, 10 and 15 Vol %) of aluminum oxide (Al_2O_3), magnesium hydroxide ($Mg(OH)_2$), silicon carbide (SiC), and hematite powder were prepared. The volume fraction of fiber, epoxy and filler materials are determined by considering the density, specific gravity and mass. Fabrication of the composites was done at room temperature by hand lay-up techniques.

2.3 Specimen Preparation

The prepared slabs of the composite materials were taken from the mold and then specimens were prepared from composite slabs for different thermal and fire resistance tests according to ASTM standards. The test specimens were cut by laminate by using different tools. Three identical test specimens were prepared for different tests.

Table 1 Designation of Composite Materials

Material Designation	Glass Fiber (%Volume)	Epoxy (%Volume)	Filler Materials (% Volume)
GE	50	50	Nil
GEA_1	50	40	10% Al_2O_3
GEA_2	50	35	15% Al_2O_3
GEM_1	50	40	10% $Mg(OH)_2$
GEM_2	50	35	15% $Mg(OH)_2$
$GESI_1$	50	40	10% SiC
$GESI_2$	50	35	15% SiC
GEH_1	50	40	10% Hematite
GEH_2	50	35	15% Hematite

3. Experimentation

3.1. Thermal Property Tests

Thermal conductivity and thermal expansion coefficient of prepared composites was determined according to standard methods.

Thermal Conductivity

Thermal conductivity measurements are carried out under steady state condition. According to ASTM E1530 disc shaped specimens with diameter of 50mm and thickness of 10mm are used in the instrument for thermal conductivity measurements. A known constant heat is applied from one side of the specimen. When the thermal equilibrium is attained and the system approaches to steady state situation, the temperature of top bottom surfaces were measured by using thermocouples installed on top and bottom of the specimen. Knowing the values of heat supplied, temperatures, and thickness, the thermal conductivity was determined by employing one-dimensional Fourier's law of conduction. All measurements are carried out approximately in the similar temperature range, i.e., 25-90 ^oC

$$Q = -KA\frac{dT}{dx} = \frac{KA(T1-T2)}{L} \qquad \text{---------- (1)}$$

Where

Q= Heat transfer rate (W)

K= Thermal conductivity (W/m ^oC)

A=Area (m^2)

T_1= temperature at bottom surface of the sample (^oC)

T_2= Temperature at top surface of the sample (^oC)

L= Thickness of the sample

dT/dx = Temperature gradient

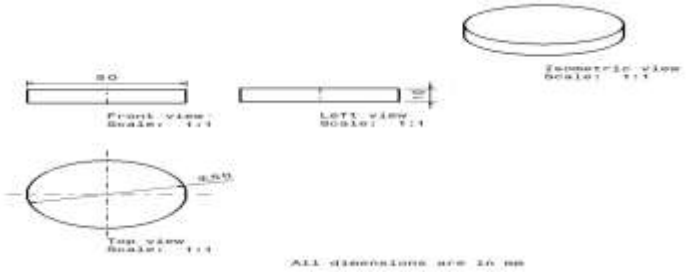

Fig. 1 Thermal conductivity test specimen

Thermal Expansion Coefficient Measurement

The specimens for the thermal expansion coefficient testing had length and thickness 90mm and 10mm respectively. The linear thermal expansion tests are performed over the temperature range of 25 to 90°C using electric furnace. The samples are slowly heated from 30 to 90 ^{0}C in the electric furnace and kept at 90^{0}C for 10 min. Thermal Expansion Coefficient is then given by the relationship

$$\alpha = \frac{\Delta L}{L \Delta T} \qquad \text{---------- (2)}$$

Where

α=Thermal Expansion coefficient
L= Original length of the sample
ΔL= Change in length of the sample
ΔT= Temperature change

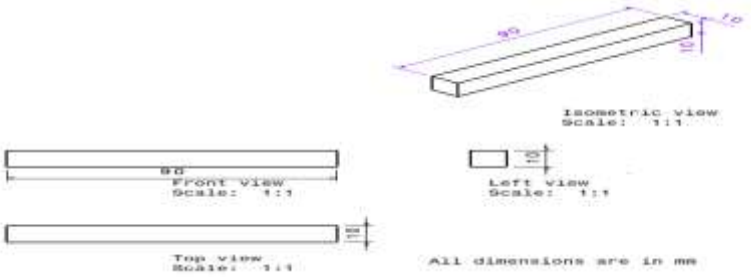

Fig. 2 Thermal expansion coefficient specimen

3.2. Fire Resistance Test

The fire properties of composite materials depend on several factors: the type of material, the fire conditions, and the test method used to measure the property.

Vertical UL94 (UL94V) Test

Rectangular shaped samples with dimensions of 127x12.7x3.2mm are exposed vertically to a methane gas burner flame as required by UL94V. The samples ignited at the bottom and burns upward. The time required for the flame to self extinguish after burner removal is measured and the occurrence of dripping on to a piece of cotton placed underneath the sample is recorded. Test is repeated for different samples. This test also classifies the materials as V-0, V-1 and V-2.
V-0 = specimens not burn more than 10 Sec and the drip do not ignite the cotton.
V-1= specimens not burn more than 30 Sec and the drip do not ignite the cotton.
V-2= specimens not burn more than 30 Sec and the drip ignite the cotton.
If the entire sample is consumed the material is classified as non-rated (NR)

Time-to-Ignition

Time to ignition is the period of time that a combustible material can with stand exposure to a constant heat flux before igniting and undergoing sustained flaming combustion, more simply, it is the time taken for a material to start burning. The ignition time can be used as a rough measure of the flammability resistance of material. Obviously it is desirable to use material with long ignition times in high fire risk applications. Extending the time- to- ignition value reduces the fire hazard of composite material used in an aircraft. The unit of time-to-ignition is seconds (Sec).

Flame Propagation Rate

The rate of the movement of the flame front is defined as the flame propagation rate.

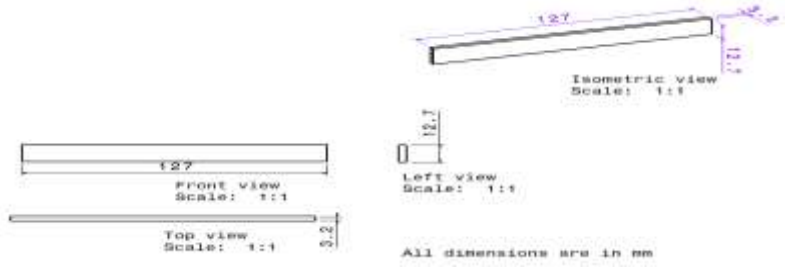

Fig. 3 UL-94 Vertical test specimen

4. Results and Discussion

The thermal conductivity, thermal expansion coefficient, time to ignition and flame propagation rate for different composition of composite materials are presented in tables 2- 5 and their variations shown in figures 4 to 7 respectively.

4.1 Thermal Conductivity

Thermal conductivity is the property describing a material's ability to transfer heat. It is well known that thermal conductivity of the composite is dependent on such factors as Polymer-filler interaction and filler characteristics, namely type and shape of filler. From the obtained results it is observed that composites filled by (10% Vol) Al_2O_3 and Mg $(OH)_2$ exhibited low thermal conductivity it may be due to that while heating of materials slow chemical reaction takes place in Al_2O_3 and Mg $(OH)_2$ and releases the small amount of water and this released water resist the heat flow and seems possible that there is a fundamental difficult in transferring heat from the matrix to the fibers. Composites filled by (10% Vol.) SiC exhibited maximum thermal conductivity (3.515 W/m ^oC). From the literature review we can observed that SiC particles having good thermal conductivity property. Hematite filled composites also exhibited high thermal conductivities when compared with Al_2O_3 and Mg $(OH)_2$ because hematite powder contains the iron particles and these particles enhances the heat transfer rate.

Table 2 Comparison of Thermal Conductivity

Composite materials	Thermal Conductivity(W/m ^oC)
GE	2.89
GEA_1	1.32
GEA_2	1.72
GEM_1	1.565
GEM_2	2.38
$GESI_1$	3.515
$GESI_2$	2.765
GEH_1	2.45
GEH_2	3.06

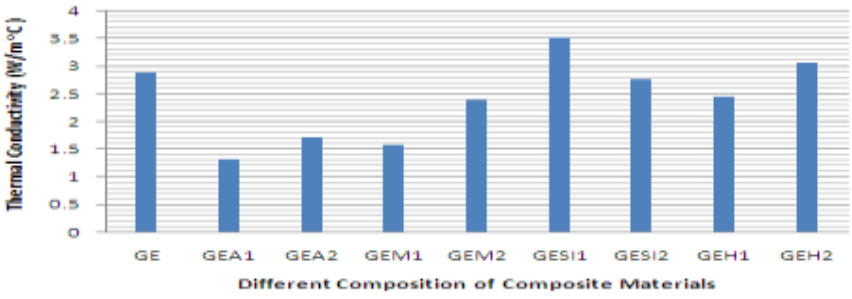

Fig. 4 Thermal conductivity for different composition of composite materials

4.2Thermal Expansion Coefficient

The experimental values of the thermal expansion coefficient of composites are presented in table-3

Table 3 Comparison of Thermal Expansion Coefficient

Composite materials	Thermal Expansion Coefficient (/^oC)
GE	1.96×10^{-5}
GEA_1	2.40×10^{-5}
GEA_2	1.66×10^{-5}
GEM_1	2.44×10^{-5}
GEM_2	1.11×10^{-5}
$GESI_1$	7.40×10^{-6}
$GESI_2$	3.70×10^{-6}
GEH_1	1.85×10^{-5}
GEH_2	1.85×10^{-5}

From the experimental results it is observed that increase the addition of Al_2O_3, Mg $(OH)_2$ and SiC to composites reduces the thermal expansion coefficient. It has been noticed that (15% Vol) Al_2O_3, Mg $(OH)_2$ and SiC filled composite exhibited less thermal expansion coefficient this may be by adding the more fillers in composite materials providing good filler matrix interaction in the system, the filler binds the matrix and prevents it from expanding as much as it would on its own. Subsequently, this would affect the thermal expansion of the composite system. The many studies have shown that materials with higher filler content leads to a lower thermal expansion coefficient. Composites filled by (15% Vol.) SiC exhibited low thermal expansion coefficient (3.70×10^{-6}) when compared with other filled composites this due to that SiC particle having reduced thermal expansion and good dimensional stability. Hematite filled composites exhibited good thermal stability.

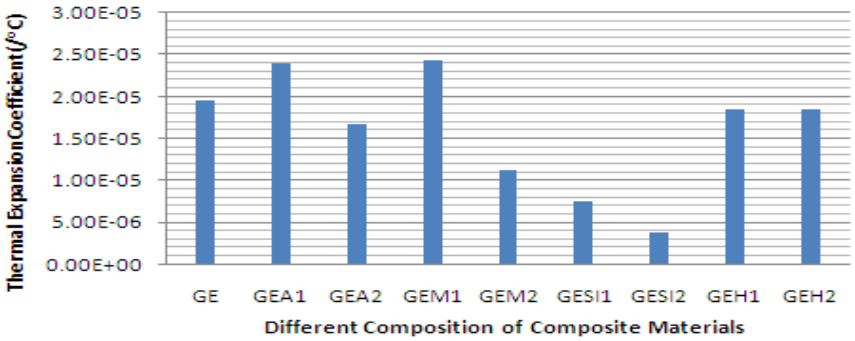

Fig.5 Thermal expansion coefficient for different composition of composite materials

4.3Time to Ignition and Flame Propagation Rate

A popular method to reduce the flammability of composites is the addition of inert fillers like silica or thermally active fillers like hydrated oxides to the polymer matrix.From the figs 6 and 7 it is observed that increase the loading of Al_2O_3, and Mg $(OH)_2$ increase the time to ignition and reduces the flame propagation rate because Al_2O_3 is active in both the condensed and gas phases of the combustion process and it is remarkably effective in suppressing flaming combustion and smoke. The main condensed phase mechanism of aluminum oxide is the absorption of heat when the filler decomposes. And this is a highly endothermic reaction that absorbs heat; another important aspect of the reaction is the creation of water vapor formed from the hydroxyl groups binded to the aluminum. This water is released into the flame where it hinders combustion by diluting the concentration of flammable gases evolved from the polymer matrix and restricting the access of oxygen to the composite surface. Magnesium hydroxide also acts as flame retardants in a similar manner to Al_2O_3 with several flame retardant mechanisms occurring in a fire. Magnesium compound undergo a highly endothermic decomposition reaction that slows the heating rate of the host material in a fire. In addition, the hydroxyl groups bonded to the magnesium are converted in the reaction into water vapor, which dilutes the concentration of flammable organic volatiles and H/Oh radicals in the flame. The decomposition of magnesium compounds also yields magnesia (MgO) that has good insulating properties. The flame retardant mechanisms of magnesium hydroxide are effective in prolonging the ignition time and reducing the amount of smoke produced by high temperature polymers. More addition of iron based hematite filler decreases the time to ignition and flame spread rate fe and derivatives are believed to decompose at elevated temperature which promotes the formation of char via a Lewis acid reaction process. The increased char yield reduces the amount of flammable organic volatiles released by decomposing polymer, which lowers the fuel load in the flame. From the obtained results we can concluded that Al_2O_3, Mg $(OH)_2$ are good flame retardant fillers. All the composites having flame extinguished time more than 30sec and failed to come under UL-94 rating.

Table 4 Comparison of Time to Ignition

Composite materials	Time To Ignition (Sec)
GE	08
GEA_1	11
GEA_2	14
GEM_1	12
GEM_2	15
$GESI_1$	09
$GESI_2$	11
GEH_1	10
GEH_2	12

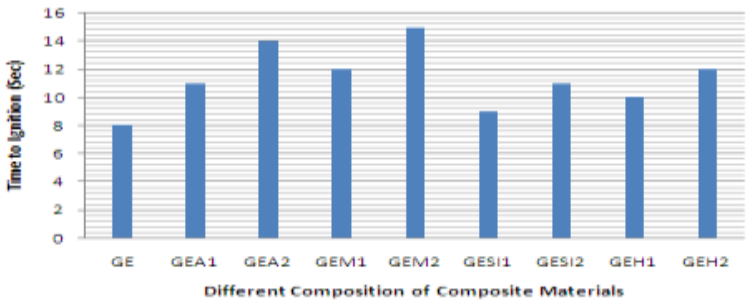

Fig.6 Time to ignition for different composition of composite materials

Table 5 Comparison of Flame Propagation Rate

Composite materials	Flame propagation Rate (mm/sec)
GE	0.60
GEA_1	0.47
GEA_2	0.44
GEM_1	0.49
GEM_2	0.45
$GESI_1$	0.512
$GESI_2$	0.52
GEH_1	0.537
GEH_2	0.514

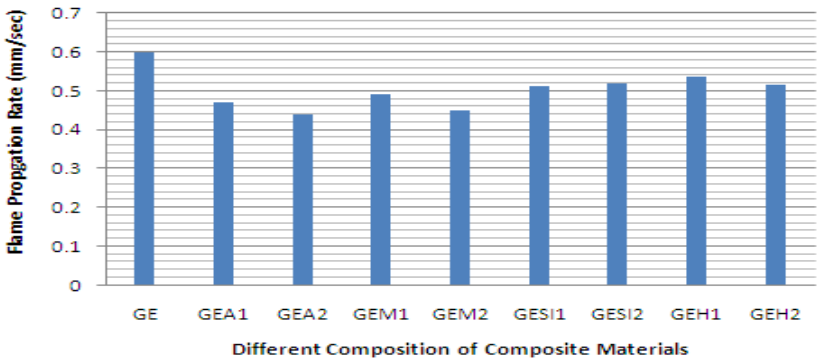

Fig. 7 Flame propagation rate for different composition of composite materials

5. Conclusions

1. From the obtained results it is observed that composites filled by (10% Vol) Al_2O_3 and Mg $(OH)_2$ exhibited low thermal conductivities. Composites filled by (10% Vol.) SiC exhibited maximum thermal conductivity (3.515 W/m ^oC). Hematite filled composites exhibited high thermal conductivities when compared with Al_2O_3 and Mg $(OH)_2$ filled composites.
2. when increase the adding of filler materials to composites reduces the thermal expansion coefficient it is seen from the composites filled by (15% Vol) Al_2O_3, Mg $(OH)_2$ and SiC when compared with (10% Vol) filled composites. Composites filled by SiC exhibited low thermal expansion coefficient when compared with other filled composites.
3. Fire test results show that increase the loading of Al_2O_3, Mg $(OH)_2$, and hematite powder increase the time to ignition and reduces the flame propagation rate of composites. From the obtained results we can concluded that Al_2O_3 and $Mg(OH)_2$ are good flame retardant fillers.
4. All the specimens having flame extinguish time more than 30 sec and failed to come under UL-94V rating.

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