

Investigation on the effect of micro and nano fillers on electrical and thermal conductivity of glass epoxy hybrid composites

Bommegowda Kabbala Basavarajappa^{1, 2*}, Nijagal Marulaiah Renukappa², Jagannathan Sundara Rajan³

¹ Department of Electronics and Communication Engineering, N.M.A.M. Institute of Technology (Affiliated to the Visvesvaraya Technological University, Belagavi) Nitte, 574110 Udupi, Karnataka, India

² Department of Electronics and Communication Engineering, JSS Science and Technology University, 570006 Mysuru, Karnataka, India

³ Department of Electrical and Electronics Engineering, Siddaganga Institute of Technology, 572103 Tumakuru, Karnataka, India

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Abstract

The electrical and thermal properties of polymer composites are enhanced by the incorporation of nano and micron fillers. Reported work on polymer composites with the combination of micro and nano sized fillers like silicon dioxide; alumina, silicon carbide, molybdenum disulphide, and graphite are limited. In this investigation, the AC conductivity of composites with the combination of micro and nano fillers was determined over a frequency range of 20 Hz to 10 MHz, at temperatures of 25, 50 and 75±2° C. The thermal conductivity of composites was also determined to investigate the synergistic effects of the hybrid fillers. The AC conductivity of the composites shows minimal dependence in the low-frequency range, and it increases at higher frequencies. Composites with hybrid fillers exhibit relatively higher AC conductivity of 10-3 S/m at 75°C. The thermal conductivity of 0.68 W/m k which is achieved with molybdenum disulphide filler, is twice the value of the base epoxy. The composites with the combination of micro and nano sized fillers also reveal good enhancement of glass transition temperature to 145°C and exhibit better electrical and thermal properties than the composites with individual micron or nano fillers.

Keywords: AC Conductivity; Epoxy Resin; Glass Woven Fabric; Micron Filler; Nano Filler; Thermal Conductivity.

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INTRODUCTION

Traditional polymer composites have serious limitations in meeting the demands of the industry and hence polymer nanocomposites were potential replacements for mainly insulation applications [1-2]. A key factor driving the increased applications of polymer composites in recent years is the innovations and advancements in the area of fiber reinforced composites. Contemporary developments in high-performance polymer resins and fiber/fabric reinforcements have evolved as a

new generation of polymers and the incorporation of nano and micro fillers have helped enormously in improving the overall characteristics of polymers. The glass reinforcements impart higher mechanical and physical properties to the polymer matrix [3]. Different fiber reinforcements namely long fibers, chopped fibers, woven mat, chopped mat are incorporated in the polymer composites to improve the mechanical and tribological properties. The properties of the composites depend heavily on the fiber alignment or the nature of the fabric used [4]. For example, glass

* Corresponding Author Email: bgowda_kbl@nitte.edu.in

fiber reinforced polymer composites have higher impact load tolerance, stiffness, and high specific strength [5]. On the other hand, E-glass fibers are preferred as reinforcement when better electrical insulating properties, low susceptibility to moisture, high mechanical properties, and low cost are the prerequisites [6].

Epoxy, polyester, and vinyl ester are the preferred polymer matrix systems for a vast variety of applications. Epoxy has excellent chemical and corrosion resistance [7]. The majority of the reported studies on polymer nanocomposites are directed towards improving the mechanical, thermal, and electrical properties in isolation. Since it is very essential to maintain a strategic balance of mechanical, thermal, and electrical properties for use under different environmental conditions, all the properties of the composites become critical for industrial applications. Epoxy composites with well-balanced physical, mechanical, thermal, and electrical properties find application in different fields. They are preferred dielectric materials for use as electrodes in electrochemical sensing applications with the incorporation of conducting fillers. For outdoor insulations applications with good tracking and erosion properties, different nano-fillers are used. Similarly, for applications involving voltage barriers, anti-corrosion coatings, aerospace structures, structural applications, and automobiles, the use of polymer composites is preferred. Thus, polymer composites are considered a key component of the present-day industry [8].

Epoxy resin is the most widely used polymer due to its excellent properties such as good compatibility with different fillers leading to improvement in the interfacial properties, higher stiffness, and better mechanical properties, reduced moisture absorption, excellent resistance to chemicals and heat. Epoxy resins are also ideal polymer matrix systems for electronics and electrical systems as well as many other industrial applications [9-10]. Epoxy resins by nature are poor conductors of heat and electricity due to their cross-linked structure. The engineering applications of epoxy resins are abundant [11] and they are increasing due to recent advances and innovations. The electrical and thermal properties of an epoxy matrix can be further enhanced by the incorporation of fillers of nanometer dimensions or by using micron sized fillers or by using a combination of nano and micron fillers.

To achieve electrically insulating and thermally conducting polymer composites, fillers such as silicon carbide, alumina, and silica are used [12]. The electrical conductivity of the fillers is a significant factor for achieving higher electrical conductivity. Electrical conductivity is observed to obey universal power-law at higher frequencies, and it minimally depends on the temperature in the lower frequency range, and the dipolar relaxation mechanisms occur in the intermediate frequency region, depending on the nature of the fillers [13-14]. The thermal conductivity of the composites is dependent on Polymer-filler interaction and filler characteristics like thermal conductivity, surface area and shape [15]. The combination of micron and nano-sized fillers in a polymeric matrix produces greater stiffness and strength in comparison to the individual fillers when used alone and the superposition of the properties of the fillers helps in enhancing the range and magnitude of the properties of the hybrid composites [16]. Though extensive literature is available on nanocomposites and micron filler-based polymer composites, there is little reported work on hybrid polymer composites using a combination of nano and micron fillers. In this investigation, different micro and nano filler particles were incorporated into the epoxy matrix which is reinforced with ECR-glass woven fabric. The effect of fillers on the AC conductivity at 25, 50 and 75° C and the thermal conductivity of the composites have been investigated and the results are discussed in this paper. In addition, the glass transition temperature T_g is also determined to understand the effect of hybrid fillers.

The selection of fillers was initially based on the good performance of the fillers in mechanical properties and their ability to improve the wear and erosion resistance. The availability of very few reported studies on the electrical and thermal properties of such composites was the motivation for this work. As a representative of the electrical engineering application, two fillers namely alumina and silica have been used while other fillers are based on better performance in wear and erosion characteristics. Fillers like graphite and SiC have been independently used as a micron or as nano fillers but the reported investigations on the hybrid combination of these fillers in a polymer matrix are extremely limited.

The objective of this study was to understand the role of incorporating small wt. % of nanofiller

Table 1. Details of resin, hardener, reinforcement, and the fillers.

Sl. No.	Materials	Source	Average fiber/nano and micron filler size	Density (g/cm ³)
1	ECR glass fabric	Owens Corning, India	18 μ m (diameter)	2.66
2	Epoxy MY740	Huntsman, USA	---	1.16
3	Hardener		---	1.2
4	Alumina	Sigma Aldrich, USA	12 nm/20 μ m	2.62
5	Silica		12 nm/25 μ m	4.0
6	Graphite		60 nm/25 μ m	2.6
7	Silicon Carbide		100 nm/25 μ m	3.1
8	Molybdenum disulphide		90 nm/20 μ m	5.06

into the glass-reinforced epoxy matrix with micron fillers. The logic behind this attempt is that the nanofillers due to their large surface area would tend to occupy the spaces between the micron fillers and thus contribute to improved filler matrix interactions. The nanofillers fillers would also help to improve the glass fiber-polymer matrix adhesion. The use of semi-conducting fillers like MoS₂, SiC, and graphite with weight percentage (wt. %), below the levels of percolation thresholds of electrical conductivity helps to increase the thermal conductivity with minimal reduction in the insulating properties. Such polymers with reasonably good insulating properties, having higher thermal conductivity find many applications in the industry. Hence the other objective of this investigation was to understand the effect of semiconducting fillers on AC conductivity and the thermal conductivity of the composites when a hybrid combination of such fillers is used.

EXPERIMENTAL METHOD

Materials

The particulars of the resin, hardener, reinforcement, and fillers are furnished in Table 1. The nano fillers of MoS₂, graphite and SiC were prepared from the micron sized filler particles with indigenous facilities.

Method of Fabrication

The hand lay-up method is the simplest fabrication technique for the preparation of

polymer composites. In this investigation, the wet hand lay-up method was employed. The ECR glass woven roving fabric was manually positioned on a porous teflon sheet. Before mixing, the epoxy resin and hardener were preheated in a vacuum chamber at 65° C for 1h 30 min to reduce the viscosity and eliminate trapped air. The epoxy resin and the hardener were then mixed in the ratio of 100 (epoxy):85 (hardener) parts by weight (pbw) as suggested by the manufacturer and the accelerator of 2.5 pbw was then added. To prepare the hybrid filler-based epoxy composites, the required weight percentage of the fillers were mixed with the epoxy-hardener mixture, using a high shear mixer which was operated at 2000 rpm for 15 min. The temperature was held at 50° C during the mixing process. The resin filler mixture was then infused onto the surface of the glass fabric which was positioned on the teflon sheet. The epoxy impregnated glass fabric was then stacked one above the other with the application of pressure using a hand roller. By this method, the trapped air bubbles and the excess of the epoxy resin was squeezed out. The procedure was repeated until the stacked layers were built up to 3 mm thickness. The translucent teflon film helped to protect the stack and a 3 mm spacer was used to ensure uniform thickness of the fabricated composites. The entire assembly was then subjected to a pressure of 0.5 MPa in a hydraulic press with bagging. The composites fabricated were pre-conditioned at a temperature of 25 ± 2° C and relative humidity of 50 ± 5 % for 24 h,

Table 2. Identification of glass epoxy composites. (Figures in parentheses show the weight percentage used)

Sl. No.	Nomenclature	Description
1	GE	ECR glass fabric (60) + epoxy (40)
2	GEA	ECR glass fabric (60) + epoxy (35) + nano alumina (1) + micro alumina (4)
3	GES	ECR glass fabric (60) + epoxy (35) + nano silica (1) + micro silica (4)
4	GESiC	ECR glass fabric (60) + epoxy (35) + nano silicon carbide (1) + micron silicon carbide (4)
5	GEM	ECR glass fabric (60) + epoxy (35) + nano molybdenum disulphide (1) + micron molybdenum disulphide (4)
6	GEG	ECR glass fabric (60) + epoxy (35) + micron graphite (4) + nano graphite (1)

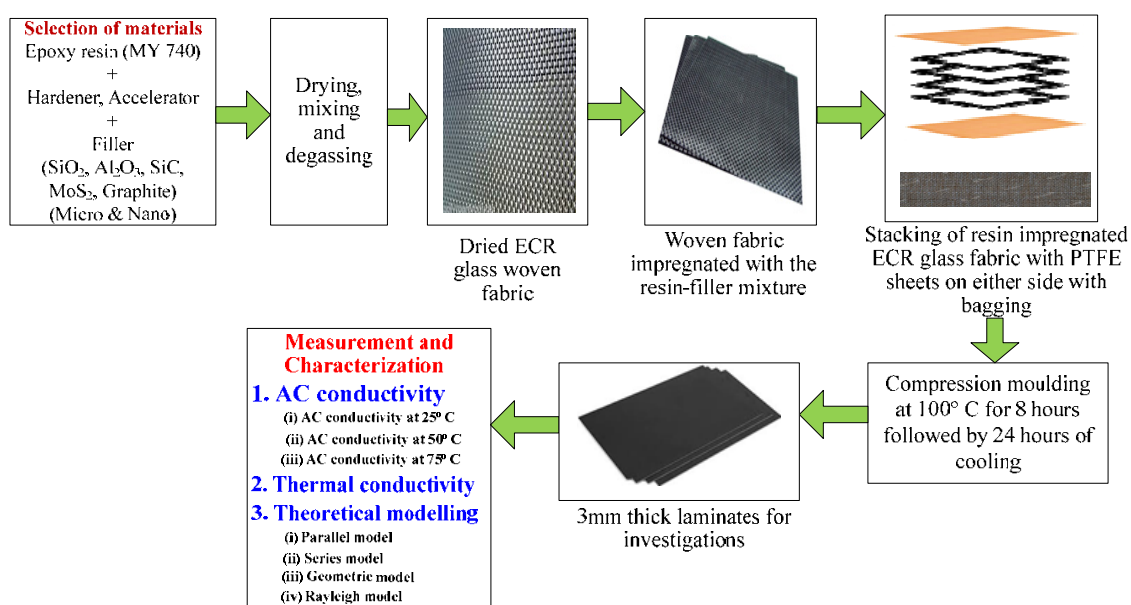


Fig. 1. Block diagram depicting an overview of the investigation.

before the commencement of the measurements. The composites fabricated were of size 250 mm x 250 mm x 3 mm. The details of composites and their nomenclature are shown in Table 2. A block diagram graphically describing the details of the materials, fabrication processes, measurements undertaken, and the parameters considered for discussions is shown in Fig. 1.

Method of Measurement

AC Conductivity

The AC conductivity of the glass epoxy hybrid composites were determined by measuring the conductance (G) using Wayne Kerr LCR meter model 6500P in accordance with ASTM D 150-11 specification.

A three-terminal guard ring electrode assembly

was used for dielectric measurements. The measuring electrode had a surface area of 20 cm². A circular specimen having a 5 cm diameter and a thickness of about 0.3 cm is placed between a pair of solid electrodes to measure the conductance (G) at an applied voltage of 1 V (rms) and the frequency is swept from 20 Hz to 10 MHz. Since the focus of the study was to understand the nature of AC conductivity, its power-law dependence and the relaxation mechanisms involved, the frequency range of 20 Hz to 10 MHz was used for measurements. The AC conductivity was measured at room temperature of 25° C, and elevated temperatures of 50° C and 75° C respectively. The temperature was maintained within ±2° C during the course of the measurements.

For high-temperature measurements, a precise

Table 3. Thermal conductivity of fiber, matrix and fillers.

Component	Thermal conductivity (W/m. K)
Fiber	1.35
Epoxy	0.24
Alumina	30
Silica	2.6
Silicon carbide	120
Molybdenum disulfide	44
Graphite	470

temperature unit with an accurate temperate controller was used. The AC conductivity (σ_{ac}) was computed from the measured parameters using the following equation:

$$\sigma_{ac} = \frac{G \times t}{A} \quad (1)$$

In this equation, G is the conductance in Siemens (S), t is the thickness of the sample in cm and A is the area of the electrode in cm².

Thermal conductivity

Thermal conductivity was measured using an Anter Unitherm TM 2022 thermal conductivity instrument, and the measurements were carried out in accordance with ASTM E1530-19. The samples were circular in shape with a diameter of about 5 cm, and a thickness of 0.3 cm. The samples were maintained at a temperature of 50° C and a compressive load of 280 kPa is applied and at thermal equilibrium, measurements are recorded. The thermal conductivity of the different components used in the hybrid composites namely fiber, matrix and fillers are tabulated in Table 3 to facilitate comparison of the thermal conductivity of the composites.

RESULTS AND DISCUSSION

Variations in AC conductivity

AC conductivity is an important property and the inclusion of semiconducting fillers like MoS₂, SiC and graphite are expected to result in much higher AC conductivity. It is also critical to examine the likelihood of percolation effects within the wt.% used. The results of the measurements at 25° C is discussed and compared with the corresponding results at 50 and 75° C, to understand the effect of temperature.

Variations in AC conductivity at 25° C

The variations in the AC conductivity (σ_{ac}) of

the composites at 25° C, is depicted in Fig. 2. Since the variations in the frequency of 20 Hz to 1 kHz were not clearly distinguishable, the variations are shown over two regions of frequency namely 20 Hz to 1 kHz and the second in the range of 5 kHz to 10 MHz. The σ_{ac} does not show significant variations from 1 kHz to 5 kHz. The changes in σ_{ac} of composites at 25° C for the frequency of 20 Hz to 1 kHz is shown in Fig. 2 (a). The results substantiate that there are minimal changes in the AC conductivity of the individual composites ranging from 4.5×10^{-9} to 1×10^{-8} S/m, from 20 Hz to 1 kHz. AC conductivity of the composite GEG is the highest at 1×10^{-8} S/m. In case of other composites, AC conductivity remains around 10^{-9} S/m, and it is not very significantly influenced by the fillers.

The variations in σ_{ac} of the composites in the frequency band of 5 kHz - 10 MHz are shown in Fig. 2 (b). In this frequency range, a linear dependence of σ_{ac} with frequency is observed and the rate of increase of σ_{ac} with frequency is observed to depend on the hybrid fillers used. The increase of σ_{ac} with frequency is seen to be maximised in GEG, and GES. This fact highlights the role of fillers in the polarization mechanisms involved. The overall variation of σ_{ac} values ranges from 1×10^{-5} to 3.2×10^{-4} S/m in the frequency band of 5 kHz - 10 MHz. The values of the AC conductivity of the composites are shown in Table 4 for a comparative evaluation of the values over the three different ranges of frequency. The results demonstrate that GEG has relatively the highest AC conductivity from 20 Hz to 5 kHz, but in the frequency range of 5 kHz to 10 MHz, the differences between the AC conductivity of all the composites except GE, are in the vicinity of 10^{-4} S/m. This is an important result which implies that though some of the fillers are semiconducting in nature; their wt. % does not result in enhancement of the AC conductivity.

Some authors [17] have reported AC conductivity of 5 to 10×10^{-3} S/m at 25° C, in the frequency band of 10 Hz -10 MHz, for nano-

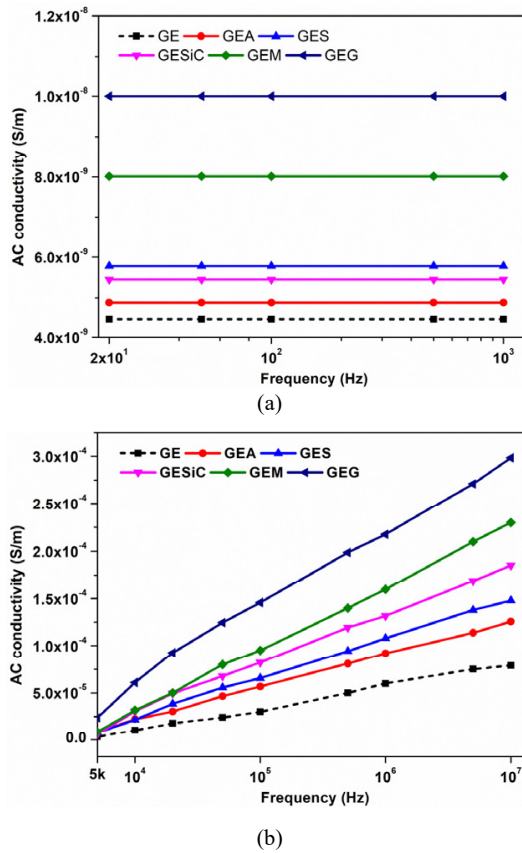


Fig. 2. Variation of σ_{ac} for GE composites at 25° C (a) 20 Hz to 1 kHz and (b) 5 kHz to 10 MHz.

Table 4. AC conductivity of the composites at 25° C in different frequency regions.

Composite	AC conductivity (S/m) at 25° C		
	20 Hz – 1 kHz	1 kHz – 5 kHz	5 kHz – 10 MHz
GE	4.45×10^{-9}	$4.45 \times 10^{-9} - 3.11 \times 10^{-6}$	$3.11 \times 10^{-6} - 7.95 \times 10^{-5}$
GEA	4.88×10^{-9}	$4.88 \times 10^{-9} - 8.27 \times 10^{-6}$	$8.27 \times 10^{-6} - 1.25 \times 10^{-4}$
GES	5.78×10^{-9}	$5.78 \times 10^{-9} - 7.83 \times 10^{-6}$	$7.83 \times 10^{-6} - 1.48 \times 10^{-4}$
GESiC	5.45×10^{-9}	$5.45 \times 10^{-9} - 6.00 \times 10^{-6}$	$6.00 \times 10^{-6} - 1.85 \times 10^{-4}$
GEM	8.01×10^{-9}	$8.01 \times 10^{-9} - 8.50 \times 10^{-6}$	$8.50 \times 10^{-6} - 2.30 \times 10^{-4}$
GEG	1.00×10^{-8}	$1.00 \times 10^{-8} - 2.34 \times 10^{-5}$	$2.34 \times 10^{-5} - 3.07 \times 10^{-4}$

alumina and nano-silica in the epoxy matrix. However, the authors have used 75 wt. % of ECR glass fiber as against 60 wt. % used in this work. The increase in σ_{ac} of the reported work of the authors is attributed to the decrease in the free space length or the interparticle filler distances, which is a direct consequence of higher wt. % of the ECR glass fiber reinforcement and reduction in volume available for the fillers to occupy. Further, the authors [17] have used only nanofillers and hence an improvement of one to two orders of magnitude of σ_{ac} is observed as compared to the

present work, at 25° C. Thus, variations in the σ_{ac} of the composites by one to two orders of magnitude are observed when hybrid fillers are used in place of nano-sized fillers.

Variations in AC conductivity at 50° C

The variations in AC conductivity of hybrid composites at 50° C is depicted in Fig. 3 (a) and (b) respectively for the two ranges of frequencies. As observed at 25° C, σ_{ac} of the composites is invariant with frequency, in the low-frequency range of 20 Hz to 1 kHz and the variations in σ_{ac}



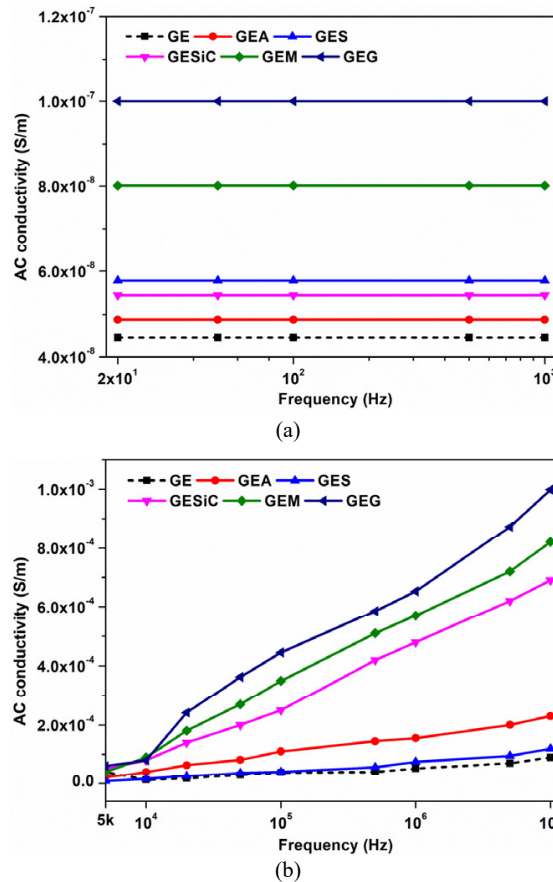


Fig. 3. Variation of σ_{ac} for GE composites at 50° C (a) 20 Hz to 1 kHz and (b) 5 kHz to 10 MHz.

Table 5. AC conductivity of the composites at 50° C in different frequency regions.

Composite	AC conductivity (S/m) at 50° C		
	20 Hz – 1 kHz	1 kHz – 5 kHz	5 kHz – 10 MHz
GE	4.45×10^{-8}	$4.45 \times 10^{-8} - 4.00 \times 10^{-5}$	$4.00 \times 10^{-5} - 9.00 \times 10^{-5}$
GEA	4.88×10^{-8}	$4.88 \times 10^{-8} - 2.07 \times 10^{-5}$	$2.07 \times 10^{-5} - 2.80 \times 10^{-4}$
GES	5.78×10^{-8}	$5.78 \times 10^{-8} - 8.05 \times 10^{-6}$	$8.05 \times 10^{-6} - 1.30 \times 10^{-4}$
GESiC	5.45×10^{-8}	$5.45 \times 10^{-8} - 3.18 \times 10^{-5}$	$3.18 \times 10^{-5} - 4.28 \times 10^{-4}$
GEM	8.01×10^{-8}	$8.01 \times 10^{-8} - 4.00 \times 10^{-5}$	$4.00 \times 10^{-5} - 8.20 \times 10^{-4}$
GEG	1.00×10^{-7}	$1.00 \times 10^{-7} - 6.05 \times 10^{-5}$	$6.05 \times 10^{-5} - 10 \times 10^{-3}$

ranges from 4.5×10^{-9} to 1×10^{-7} S/m. Thus the AC conductivity values at 50° C shows an increase by one order of magnitude as compared to the values at 25° C. The effect of fillers at 50° C is seen to minimally improve the σ_{ac} of the composites GES and GEA over that of GE. The marginal difference can be visualized from the data of AC conductivity over a different range of frequencies as shown in Table 5. The highest σ_{ac} is observed in case of GEG ($=1.00 \times 10^{-7}$ S/m) and in this aspect, the results are similar to the trends observed at 25° C. The overall variations in σ_{ac} of the other composites is

observed to be around 10^{-8} S/m.

A comparison of the low-frequency AC conductivity data at 25 and 50° C (Table 4 and 5) reveals a uniform one power increase of σ_{ac} due to an increase in temperature. As observed at 25° C, all the composites individually depict a near frequency invariant AC conductivity in the low frequency range. In the high frequency range, the rate of increase of AC conductivity with frequency is strongly dependent on the filler used.

The σ_{ac} of the composites at 50° C in the frequency range of 1 kHz to 5 kHz varies from 10^{-8}

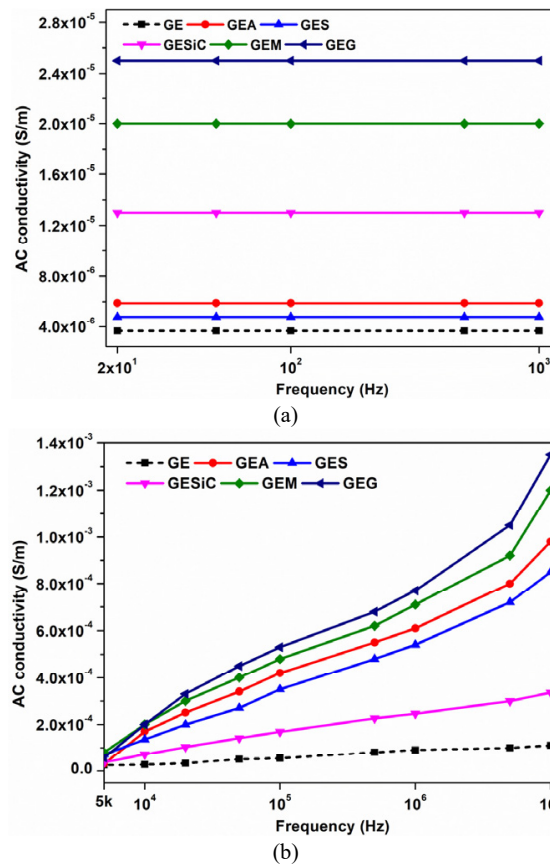


Fig. 4. Variation of σ_{ac} for GE composites at 75° C (a) 20 Hz to 1 kHz and (b) 5 kHz to 10 MHz.

to 10^{-5} S/m. A one power difference is seen at 1 kHz but at 5 kHz, the difference is not incredibly significant. In the high frequency range of 5 kHz to 10 MHz, the overall variations in σ_{ac} is from 10^{-5} to 10^{-3} S/m. It is important to note that in GE, σ_{ac} shows minimal improvement as compared to the corresponding value in the lower frequency range of 1 kHz to 5 kHz. But in other composites, a one to two power increase in σ_{ac} is observed at 10 MHz.

A comparison of AC conductivity at 25 and 50° C shows that in the frequency range of 20 Hz-1 kHz, the increase is by one to two powers, and the corresponding increase in the frequency range of 1 kHz to 5 kHz is by one power and from 5 kHz to 10 MHz, it is by one to two powers.

Variations in AC conductivity at 75° C

The effect of hybrid fillers on the σ_{ac} of the composites at 75° C is depicted in Fig. 4 (a) and (b) respectively. There is marginal improvement in the σ_{ac} of the composites due to the increase in temperature to 75° C. The σ_{ac} of the composites

range from 4×10^{-6} to 2.4×10^{-5} S/m over 20 Hz to 1 kHz. Thus, improvement by two orders in σ_{ac} of the composites is observed with an increase in temperature from 50 to 75° C. Further, frequency invariance of σ_{ac} of the composites is observed in all the composites as observed at 25 and 50° C. The result confirms that the basic nature of the frequency response of AC conductivity of GE is retained by all the composites, and the role of the fillers on AC conductivity is seen in terms of one or two orders of increase in magnitude. Further, frequency invariance of AC conductivity is extended from 1 kHz to 5 kHz with the increase in temperature, and this feature is different from the linear increase observed at 25 and 50° C. The variations in AC conductivity of the composites over different ranges of frequency is shown in Table 6.

In the 20 Hz to 1 kHz range, GE, GEA, GES show improvement in AC conductivity by two orders as compared to the corresponding value at 50° C. However, three orders of improvement in AC

Table 6. AC conductivity of the composites at 75° C in different frequency regions.

Composite	AC conductivity (S/m) at 75° C		
	20 Hz – 1 kHz	1 kHz – 5 kHz	5 kHz – 10 MHz
GE	3.65×10^{-6}	$3.65 \times 10^{-6} - 2.50 \times 10^{-5}$	$2.50 \times 10^{-5} - 1.10 \times 10^{-4}$
GEA	5.88×10^{-6}	$5.88 \times 10^{-6} - 3.00 \times 10^{-5}$	$3.00 \times 10^{-5} - 9.80 \times 10^{-4}$
GES	4.78×10^{-6}	$4.78 \times 10^{-6} - 7.00 \times 10^{-5}$	$7.00 \times 10^{-5} - 8.50 \times 10^{-4}$
GESiC	1.30×10^{-5}	$1.30 \times 10^{-5} - 2.39 \times 10^{-5}$	$2.39 \times 10^{-5} - 3.36 \times 10^{-4}$
GEM	2.00×10^{-5}	$2.00 \times 10^{-5} - 8.00 \times 10^{-5}$	$8.00 \times 10^{-5} - 1.20 \times 10^{-3}$
GEG	2.50×10^{-5}	$2.50 \times 10^{-5} - 5.64 \times 10^{-5}$	$5.64 \times 10^{-5} - 1.35 \times 10^{-3}$

conductivity are observed in case of GESiC, and GEM and two orders of increase in AC conductivity is observed in case of GEG when the temperature is increased from 50 to 75° C.

In the frequency range of 1 kHz to 5 kHz, there is no change in the order of magnitude of the AC conductivity, and it remains between 10^{-6} to 10^{-5} S/m. Thus, the effect of temperature is not significant in influencing the AC conductivity over the frequency range of 1 to 5 kHz. In the frequency range of 5 kHz to 10 MHz, divergent trends are seen. One order of increase of σ_{ac} is seen in case of GE, GEA, GES, and GESiC and two orders of increase is seen in GEM and GEG. Thus, the effect of temperature is relatively profound in composites with semi-conducting fillers like MoS₂ and graphite. Though SiC is also semiconducting, the resulting increase in AC is lower as compared to MoS₂ and graphite filler when temperature is increased from 50 to 75° C.

The variations in AC conductivity with temperature can be understood by considering the mechanism of charge transfer. Hopping conduction would take place between the adjacent filler particles because of the localization of charge carriers in the composites. Thus, an increase in the σ_{ac} of the composites is observed at higher frequencies [13], but the increase is by only one to two orders of magnitude, signifying that the density of states of the charge carriers is not significantly higher due to the increase in temperature. The increase in AC conductivity is more significant in GEG than the other composites owing to the higher conductivity of graphite fillers.

The variations in σ_{ac} of the hybrid composites at three different temperatures indicate that σ_{ac} is temperature-dependent in the higher frequency band of 5 kHz -10 MHz. The effect of temperature resulting in the increase of σ_{ac} is ascribed to the increase in the mobility of the charge carriers within the polymer composites [13]. The electrical conduction occurs at a lower filler density in

composites based on the tunnelling effect. Accordingly, in an insulating matrix, the fillers would not make a perfect network. As a result, electrons find it difficult to travel freely between the filler particles and hence would attempt to cross the potential barrier. The fillers can permit tunnelling when they are in close or direct contact with each other or form a good network, which is possible only above the percolation density of fillers. Only under such circumstances, higher AC conductivity of the composites is possible [18-19]. Though graphite has relatively the highest electrical conductivity, the resulting increase in σ_{ac} of the composite is not very substantial as compared to other fillers which individually possess lower electrical conductivity, mainly due to the resulting band structure and the placement of fillers, which makes both hopping and tunnelling mechanisms difficult. Thus, it is evident the wt. % of the fillers used is below the respective levels for the percolation threshold of electrical conduction, even though they are semiconducting in nature.

Power Law of AC conductivity

The AC conductivity variations with frequency are associated with a factor called critical frequency f_c . This frequency is always identifiable as the frequency at which the σ_{ac} starts increasing. At f_c , there is a clear demarcation between the frequency invariant AC conductivity and the frequency dependent AC conductivity. Beyond the critical frequency f_c , the σ_{ac} of the composites exhibit the power-law dependence which follows the equation:

$$\sigma_{total} = \sigma_{dc} + A f^s \tag{2}$$

Where σ_{total} is total conductivity of the sample, σ_{dc} is DC conductivity and the term Af^s represents the AC conductivity. In this equation, 's' is the power-law exponent (generally $0 < s < 1$) and 'A' is the pre-exponential constant.

Andrew K Jonscher [20] has stated that at higher temperatures, polymers exhibit increased mobility of ions, which are either intrinsic to the lattice, as in case of ionic solids, or extrinsic, as in case of strictly covalent lattices, such as polymers, due to the presence of impurities. Some authors [21] have reported that the conductivity increases as a power of frequency with power exponent 's' at higher frequencies nearing the value of 1. Reports in the literature have shown agreement with this finding [22-23].

Anindya Sundar Das and co-authors [22] have reported that temperature-dependent and non-zero values of 's' are caused by the short-range cumulative motion of electrons or small polarons. The authors have also reported that with increasing temperature, there is an increase in the power-law exponent values, indicating the possibility of tunnelling due to the non-overlapping of small polarons. According to the authors, a high order of local lattice distortion is caused by the accumulation of charge carriers or electrons at certain locations, which activate the formation of small polarons.

The electrical conductivity of the polymer is primarily due to electron hopping. When an electric field is applied, the charges will move to add to the electrical conductivity of the polymer. The degree of contact between mobile ions and lattice around them is represented by the exponent 's' ($0 < s < 1$), and 'A' is a constant that defines the strength of the polarizability [24]. The power-law variation is characterized by: (i) a plateau region at low frequencies which correspond to σ_{dc} . In this region of frequency, the σ_{ac} (which is equal to σ_{dc}) increases with an increase in temperature but is invariant with frequency. This behaviour indicates that the electrical conductivity of the polymer is a result of the thermal activation of the charge carriers. However, the increase in σ_{ac} is in the range of two to three powers over the temperature range studied, shows that the effect of thermal activation is restricted. (ii) At higher frequency, the conductivity is directed by the second term of the Power law equation. The cause of the frequency dependence of conductivity, according to Jonscher, is the relaxation phenomenon of charge carriers which are mobile when they are in the ionic state.

The critical frequency of the composites is observed to be 1 MHz at 25, 50 and 75° C. Since most of the reported work on AC conductivity has

considered variations in the frequency band from 10 Hz - 10 MHz, from the results it appears that σ_{ac} is nearly frequency invariant over a broadband of frequency from few Hz to MHz. In order to highlight this fact, the frequency variations in AC conductivity have been discussed over two separate ranges of frequency. The AC conductivity of the individual hybrid composites is observed to increase marginally in the frequency band of 20 Hz to 1 kHz, and the rise is also minimal from 10 kHz to 1 MHz. However, it is relatively more significant in the range of 1 MHz -10 MHz. The AC conductivity values depicted in Fig. 2, 3 and 4, bring out the differences in the AC conductivity in the lower range of frequency, which is perceptible, though the magnitude of the differences is minimal. This investigation has highlighted the fact that in the low frequency range, though all composites reveal a near frequency invariant AC conductivity, though the fillers definitely influence the individual values. The differences in AC conductivity in the low frequency range, though small is not highlighted in the literature. However, these small differences arising due to the incorporation of different hybrid fillers has been identified. Thus, the important contribution of this work lies in the fact that it has been possible to establish that each hybrid filler has its own influence on the AC conductivity in the low frequency range, though it is frequency invariant. The resulting differences in the AC conductivity values at all three measurement temperatures due to the incorporation of both insulating and semiconducting fillers has been established over the frequency range of 20 Hz to 10 MHz. Such an approach has not been highlighted in the literature.

Some authors have modified the Jonscher Power-law dependence into an equation consisting of two exponents to account for the different types of conductivity mechanisms involved [25]. This equation has the form:

$$\sigma_{total} = \sigma_{dc} + Af^{s_1} + Bf^{s_2} \quad (3)$$

The two exponents are introduced to aid better interpretation of the differences in σ_{ac} in the two regions of higher frequency, and also to interpret the invariance of σ_{ac} in the lower frequency region. The exponent 's₁' is assigned to the frequency region beyond the region of frequency invariance (traditionally considered as equal to DC conductivity) and translational

Table 7. Power-law exponent of the σ_{ac} of the composites.

Composite	Value of exponent 'S' at		
	25° C	50° C	75° C
GE	0.97	0.98	0.99
GEA	0.98	0.99	0.99
GES	0.98	0.99	0.99
GESiC	0.94	0.95	0.96
GEM	0.95	0.96	0.97
GEG	0.94	0.96	0.97

ion hopping and exponent 's₂' is assigned to the higher frequency region and is an indication of the existence of localized relaxation/re-orientational mechanisms.

The Power law of equation 3 was used for the two regions of AC conductivity of the composites investigated namely 5 kHz to 1 MHz and 1 MHz to 10 MHz, at the three temperatures. Though the AC conductivity variations with frequency visually appear to follow the power-law with two different exponents s₁ and s₂ as stated in equation 3, it was not so in terms of the exponent. The AC conductivity variations of Fig. 2 (b), 3 (b), and 4 (b) indicate the probable existence of more than one electrical conduction mechanism and this is more dominantly perceptible in GESiC, GEM and GEG, which are all semiconducting in nature. Surprisingly, the values of s₁ and s₂ in the two frequency regions did not show much difference. Hence, due to the lack of sufficient evidence for the existence of different mechanisms of electrical conduction, only one exponent value representing the entire high-frequency region is considered. Thus holistically, the composites follow the principle of the Power-law dependence of σ_{ac} which is enumerated in equation 2. The indications about the existence of the Power-law dependence of equation 3 may perhaps become more evident at higher wt. % of the fillers or at temperatures above 75° C. This fact supports the view that the electrical percolation levels are not reached with the wt. % of the fillers used.

In general, there are three charge transport mechanisms. These are attributed to the following mechanisms: (i) Adjacent to the conduction or valance bands, excited charge carriers are transported to an extended state. (ii) Charge carriers are excited into localized positions at the edges of the valence or conduction bands, permitting their transport. (iii) Hopping transport by carriers with energies close to the Fermi level. The resulting relaxation is due to Quantum

Mechanical Tunnelling (QMT) because σ_{ac} is linearly dependent on temperature and the exponent is frequency-dependent and temperature-independent [21, 26]. Since this characteristic is not established over the three temperatures and frequency range used, it is difficult to attribute QMT to the observed variations.

Correlated Barrier Hopping (CBH) is another relaxation mechanism [26, 27], and it is explained as due to the electron hopping over the potential barrier among two sites and the σ_{ac} is attributed to the hopping among the defect states. Power law exponent (s) of σ_{ac} in the lower frequency regime is acknowledged to decrease with the rise in the concentration of filler. Since the values of the exponent, s₁ and s₂ did not show significant differences, the existence of CBH cannot be substantiated. Hence universal Power-law with only one exponent is considered more appropriate to account for the AC conductivity of the hybrid composites, for the wt. % of the fillers used.

The Power law exponents of the composites are shown in Table 7. It is also important to observe that, the σ_{dc} which is referred to as being equivalent to the frequency invariant σ_{ac} in the low-frequency region would not be a correct representation of the fact. Perhaps, in the exceptionally low regions of frequency, much below 20 Hz, (perhaps in the mHz range) the frequency invariant σ_{ac} would match with the σ_{dc} of the composites.

The investigations of σ_{ac} for hybrid composites, follows the universal Power-law, with an exponent value of 0.9. A marginal increase in the value of the exponent 's' is observed in the case of GESiC, GEM and GEG. The AC conductivity appears to be driven by the mobility of free charges with the increase in temperature. For further insights into the mechanism of AC conductivity of hybrid composites, higher wt. % of the fillers, closer and beyond the percolation threshold of electrical conductivity would be very useful.

Table 8. Experimental Thermal conductivity of the composites.

Composite	Experimental (W/m. K)
GE	0.28
GEA	0.38
GES	0.43
GESiC	0.52
GEM	0.68
GEG	0.54

The results of AC conductivity have established that though semiconducting fillers like SiC, MoS₂ and graphite are used, the wt. % of fillers correspond to levels below the percolation threshold for electrical conduction. Contrary to the expectations of two exponents to account for the AC conductivity mechanism, a Power law with a single exponent has been established to satisfactorily explain the AC conductivity mechanism over the entire high-frequency range. Thus, even with the use of more conducting fillers, it has been possible to restrict the AC conductivity, and the difference between the AC conductivity of the composites with traditional insulating fillers like alumina and Silica with composites consisting of SiC, MoS₂ and graphite is not remarkably different.

Thermal conductivity and its Theoretical modelling for hybrid composites

The investigations on AC conductivity of the composites have clearly established the differences due to the incorporation of the fillers. The differences in AC conductivity between the insulating and semiconducting fillers are not very apparent. However, the thermal conductivity of the composites using semiconducting fillers is expected to be much higher than the composites with insulating fillers. The thermal conductivity was experimentally determined through the thickness direction and the results are shown in Table 8. Since the composites are intended for barrier applications, the thermal conductivity through the thickness direction was determined. For further insights into the role of fillers in the thermal conductivity mechanisms, analytical prediction models were used. The models considered are the parallel, series, geometric and Rayleigh models. These models help to understand the role of each component of the polymer composite since considerable differences exist in the thermal conductivity of the epoxy matrix, glass fiber and fillers.

From the results of Table 8, it is observed that the thermal conductivity of the composite GE is 0.28 W/m. K. The increase in thermal conductivity of the composites with fillers in comparison to the composite GE shows that a 35.7 % increase results due to alumina filler, which has the excellent ability for control of fire and flame retardance. With silica, the composite GES shows a 53.57 % increase, while the corresponding increase in GESiC is 85.7 % and it is a 142.85 % increase in GEM and 92.85 % in GEG. Thus, the semiconducting fillers help in significant improvement of the thermal conductivity of the composites in comparison to the conventional fillers like alumina and silica. The composites GESiC, GEM and GEG show higher thermal conductivity and can thus be used for applications requiring higher thermal conductivity, with less stringent requirements of dielectric strength.

The reports on the theoretical estimation of thermal conductivity of the composites consisting of nano filler or micron fillers have been discussed in the literature [28]. However, the thermal conductivity of composites with the hybrid combination of nano and micron fillers are not discussed much in the literature, mainly due to the difficulties in accounting for the contributions from the nano and micron fillers. Hence this investigation attempts at the theoretical estimation of thermal conductivity by modification of the existing Models, by the introduction of constants which are modified to account for the effect of the hybrid fillers.

Poornima and co-authors [29] have estimated the thermal conductivity of the composites along the fiber direction and through the thickness. However, in the present investigation which targets barrier applications of the composites, the main focus was to understand the effect of hybrid fillers on the thermal conductivity through the thickness. The authors have also used similar models to compute the thermal conductivity of the composites and the changes in the constants

Table 9. Values of the constants of the modified Parallel model of Thermal conductivity.

Constant	Modified Parallel model					
	GE	GEA	GES	GESiC	GEM	GEG
A	0.25	0.4	0.33	0.52	0.78	0.2
B	1	0.025	1	0.025	0.02	0.02

of the model arising due to the use of carbon fillers like MWCNT and graphene nanoplatelets has been discussed. In multicomponent systems, it becomes extremely difficult to estimate the thermal conductivity and the need for changes in the existing thermal conductivity models is inevitable.

By the iterative method, different values of the constants have been estimated and the best fit values of the constants have been determined. A comparison of the results of theoretically predicted thermal conductivity with the experimental results is made for arriving at the efficacy of the proposed models.

Parallel Model

The parallel model is the simplest model for estimating the thermal conductivity of polymers by considering the volume fraction and thermal conductivity [30] and it is given by the equation:

$$k_{eff} = (k_g * v_g) + (k_m * v_m) + (k_f * v_f) \quad (4)$$

In this equation, k_{eff} , k_g , k_m and k_f are the thermal conductivity of the composite, glass fiber, matrix and fillers, v_g , v_m and v_f are the volume fraction of glass fiber, matrix and fillers. The third term of equation 4 involves the thermal conductivity of the hybrid nano and micron filler, which can be assumed to be the same. But the volume fraction of the nano and micron fillers are differ and hence to account for the volume fraction of the two fillers, two constants are introduced. The first constant accounts for the interaction between the fiber and the fillers which influences the adhesion of the epoxy to the fillers, and the constant B accounts for the contributions arising from the use of nano and micron fillers. Thus, the basic equation of the parallel model has been modified as follows:

$$k_{eff} = A (k_g * v_g) + (k_m * v_m) + B (k_f * v_f) \quad (5)$$

For the composites, the value of 'A' is observed to range from 0.2 to 0.78 and constant

'B' is in the range from 0.02 to 1. The values of 'A' and 'B' of the modified parallel model of the thermal conductivity of the composites are shown in Table 9. The variations in the value of the constant A shows that the interactions of the fillers with the glass fiber alters the contribution of the glass fibers to the total thermal conductivity of the composite, except in case of the composite GEG. It can also be due to the fact that the wt. % of graphite is insufficient for enhancement of the thermal conductivity.

Series Model

The series model is another simplest model for estimating the thermal conductivity of composites. This model considers the volume fraction and the thermal conductivity of the components of the composites [30] and it is given by the equation:

$$k_{eff} = \frac{1}{\left(\frac{v_g}{k_g}\right) + \left(\frac{v_m}{k_m}\right) + \left(\frac{v_f}{k_f}\right)} \quad (6)$$

The basic series model shown in equation 6 is revised for making this equation suitable for hybrid fillers with the introduction of the constant 'A' and the modified equation is as follows:

$$k_{eff} = \frac{1}{\left(\frac{v_g}{k_g}\right) + A \left(\frac{v_m}{k_m}\right) + \left(\frac{v_f}{k_f}\right)} \quad (7)$$

The value of the constant A is attributed to the changes that the polymer matrix undergoes due to the addition of fillers. The values of the thermal conductivity of the composites are shown in Table 10 and it is in the range of 0.7 to 1.6. The results confirm that the effect of fillers on the polymer matrix is minimal due to the incorporation of graphite, but it is relatively more significant with the other fillers.

Geometric Model

Another model to predict the effective



Table 10. Values of the constants of the modified Series model of Thermal conductivity.

Constant	Modified Series model					
	GE	GEA	GES	GESIC	GEM	GEG
A	1.5	1	0.9	0.7	1.6	1.4

Table 11. Values of the constants of the modified Geometric mean model of Thermal conductivity.

Constant	Modified Geometric mean model					
	GE	GEA	GES	GESIC	GEM	GEG
A	0.52	0.65	0.78	0.85	0.7	1
B	1	1	1	1	1	0.9

thermal conductivity of the hybrid composites is the Geometric mean model [31] and it follows the equation:

$$k_{eff} = (k_g)^{v_g} * (k_m)^{v_m} * (k_f)^{v_f} \tag{8}$$

The geometric mean model shown in equation 8 is improved for hybrid fillers with two constants A, and B respectively as shown in the equation:

$$k_{eff} = A(k_g)^{v_g} * (k_m)^{v_m} * B(k_f)^{v_f} \tag{9}$$

As in the parallel model, a constant is introduced to account for the thermal conductivity contribution from the glass fiber and another constant for the filler. The value of constant 'A' which is related to the glass fiber contributions towards thermal conductivity is observed to range from 0.65 to 1 and the value of constant 'B' which is associated with the contributions from the fillers, is in the range from 0.9 to 1.0. The values of 'A' and 'B' of the modified Geometric mean model are shown in Table 11.

With graphite, the contribution from the filler is reduced, but it helps in improving the matrix-glass fiber interactions, in a much better manner than in other composites. Thus, the model helps in identifying the dominant factor, whether it is the contribution from the fillers or the effect of filler in the fiber-polymer matrix interactions, which is responsible for the improvement in the thermal conductivity.

Rayleigh model

Another convenient way of predicting thermal conductivity is based on the Rayleigh model [32]. Prediction of the thermal conductivity based on the Rayleigh model is based on the following equation:

$$k_{eff(m,f)} = k_m \left[1 + \frac{3*v_f}{\left(\frac{k_f - 2k_m}{k_f - k_m}\right) - v_f + 1.569 \left(\frac{k_f - k_m}{3k_f - 4k_m}\right) * v_f^{\frac{10}{3}}} \right] \tag{10}$$

$$k_{eff(g,m,f)} = k_m \left[1 + \frac{2*v_g}{C_1 - v_g + C_2 (0.30584*v_g^4 + 0.013363*v_g^8)} \right] \tag{11}$$

The prediction and estimation of the thermal conductivity of the composites consisting of a polymer matrix with fillers are based on equation 10. The higher-order terms of equation 10 are neglected due to their minimal contributions. However, equation 11 is extended for the prediction of the thermal conductivity of composite consisting of the polymer matrix, glass fiber reinforcement and the filler. Equation 11 incorporates the effect of glass fibers of cylindrical shape. This model assumes that the cylindrical fibers are aligned in the polymer matrix and thus thermal conductivity of the polymer matrix with fillers and the glass fibers are considered in this model.

In equation 11,

$$C_1 = \frac{k_g + k_{eff(m,f)}}{k_g - k_{eff(m,f)}}$$

$$C_2 = \frac{k_g - k_{eff(m,f)}}{k_g + k_{eff(m,f)}}$$

The Rayleigh model of equation 11 has been modified with the introduction of constant 'A' to determine the effective thermal conductivity of the composites and it has the form:

$$k_{eff(c)} = k_m \left[1 + \frac{2*v_g}{(A*C_1) - v_g + C_2 (0.30584*v_g^4 + 0.013363*v_g^8)} \right] \tag{12}$$



Table 12. Values of the constants of the modified Rayleigh model of Thermal conductivity.

Constant	Modified Rayleigh model					
	GE	GEA	GES	GESiC	GEM	GEG
A	3.33	1.66	1.25	0.9	0.76	0.86

Table 13. Comparison of values of Thermal conductivity of the composites based on different models.

Composite	Thermal conductivity (W/m. K)				
	Prediction models				Experimental
	Parallel model	Series model	Geometric mean model	Rayleigh model	
GE	0.28	0.26	0.27	0.29	0.28
GEA	0.38	0.39	0.37	0.38	0.38
GES	0.41	0.43	0.43	0.43	0.43
GESiC	0.52	0.53	0.52	0.51	0.52
GEM	0.68	0.68	0.68	0.68	0.68
GEG	0.54	0.53	0.55	0.54	0.54

Table 14. Thermal conductivity and glass transition temperature of the composites.

Composite	Thermal conductivity (W/m. K)	Glass transition temperature (T _g) (° C)
GE	0.28	137.0
GEA	0.38	135.5
GES	0.43	135.3
GESiC	0.52	146.2
GEM	0.68	144.9
GEG	0.54	142.9

The values of the constants for the composites are shown in Table 12. It is observed that the value of 'A' is in the range of 0.76 to 3.33. This is a more comprehensive model, which shows that the constant 'A' depends on the thermal conductivity of the glass fiber and the effective thermal conductivity of the matrix with the fillers. A high value of 'A' would obviously reduce the thermal conductivity of the composite. Assuming the effect of glass fiber on thermal conductivity remains unchanged, it is observed that the incorporation of fillers tends to help in the reduction of the value of constant A. In this aspect, MoS₂ filler has the maximum impact of reducing the value of the constant.

The values of thermal conductivity of the composites based on different models are shown in Table 13. From the results, it is observed that there is a good correlation between the predicted and experimental values of the thermal conductivity.

The thermal conductivity of epoxy with unidirectional glass fibers is 0.2 W/m. K [33], whereas the thermal conductivity of epoxy with woven glass fabric (GE) is 0.28 W/m. K.

A comparison of the thermal conductivity of individual components of the composites and the thermal conductivity of the hybrid composites shown in Table 3 and Table 8 reveals that the fillers with higher thermal conductivity help in the enhancement of the thermal conductivity of the composites. The values of thermal conductivity of this investigation are 10 to 15 % higher than the value reported by Suchitra and co-authors [28] for nanofillers.

Having established the thermal conductivity, it is also important to investigate the effect of hybrid fillers on the T_g of the composites. The glass transition temperature and thermal conductivity of epoxy and epoxy with hybrid fillers are tabulated in Table 14 for comparison. The T_g of the composites GEA and GES is reduced minimally by 1.6 and 1.7° C and considering the possibility of error in temperature and the instrument or measurement errors, it can be presumed that alumina and silica in hybrid combination do not affect the T_g of the epoxy composite. However, their thermal conductivity values are relatively higher than the GE composite. In case of GESiC, the increase in T_g is by 9.1° C, while it is by 7.8°

C in case of GEM and in case of GEG it is by 5.84° C. These composites show much higher thermal conductivity than the GE composite. Thus, the semiconducting fillers help to increase the thermal conductivity and T_g of the composites.

Nanostructures in polymers have unveiled new perceptions of true multifunctional composites. MoS_2 , SiC and graphite have the potential to reach high orders of electrical conductivity and thermal conductivity. However, the wt. % of the hybrid fillers and the ratio of nano to micron sized fillers is crucial for the enhancement of thermal properties. The thermal conductivity though might increase with wt. % of fillers up to a certain level, but it is the size of the filler which is crucial. The variations in the thermal conductivity of composites with different semiconducting fillers imply that there is some other mechanism coming into the picture, other than the movement of electrons as in the case of electrical conductivity.

The mechanism of thermal conductivity of hybrid epoxy composites is a complex phenomenon. Each filler, has unique characteristics and when it is incorporated into the polymer, advantages and some limitations of the fillers would result. The interfacial regions scatter phonons and thus the heat absorbed is not transferred through epoxy-filler interfaces. The interfaces become more complex due to the variations in the size and distribution of nano and micron fillers. Heat is transferred from one side of the filler to the other side within the polymer, and it passes through a noticeably shorter distance due to the scattering of the phonons. In a polymer composite, fillers like SiC, MoS_2 and graphite tend to form a thermal conductive channel which helps phonon transfer from the fillers to the polymer and from the polymer to the filler [34]. It is also likely that the molecular chains of epoxy and the molecular chains on the surface of fillers intertwine to form an interlayer [35].

CONCLUSION

In this investigation, the AC conductivity and thermal conductivity of epoxy composites with the combination of micron and nano sized fillers has been investigated. Based on the experimental results, the following conclusions are drawn:

(i) AC conductivity of the hybrid composites is frequency invariant from 20 to 1 kHz and increases marginally from 5 kHz to 5 MHz and more significantly from 5 to 10 MHz.

(ii) The highest σ_{ac} is observed in case of GEG at 25, 50 and 75° C and the increase in σ_{ac} over a temperature range of 25 to 75° C is from 3×10^{-4} to 1.4×10^{-3} S/m.

(iii) At 25 and 50° C, the trends in the variations of σ_{ac} with frequency remains identical and only a marginal increase in the magnitude of σ_{ac} with temperature is observed from 25° C to 50° C.

(iv) The highest increase in σ_{ac} of the composites is observed over the frequency range of 1 MHz to 10 MHz at 75° C.

(v) The Power law exponent shows only a minimal increase with temperature in GEG and GESiC, and it is invariant with temperature in other composites, demonstrating the fact that not all hybrid fillers show identical trends in AC conductivity.

(vi) From the AC conductivity and its Power-law dependence, 1 MHz is identified as the critical frequency.

(vii) The thermal conductivity of the hybrid composites GESiC and GEG is 1.9 times the value of the base epoxy and in GEM, it is 2.4 times the value of GE. Thus, semiconducting fillers show a much higher increase in thermal conductivity than the increase seen in AC conductivity.

(viii) The composites with hybrid semiconducting fillers show good enhancement of the T_g varying from 6 to 9° above that of GE. Thus, improvement in thermal stability with semiconducting filler is demonstrated.

(ix) Fillers like silica and alumina show a minimal increase in thermal conductivity as compared to semiconducting fillers.

(x) The role of hybrid fillers is distinct in the frequency response of AC conductivity and magnitudes of the thermal conductivity, and this fact implies that the role of fillers in the two mechanisms responsible for electrical conduction and phonon transfer is different.

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CONFLICT OF INTEREST

Authors have no conflict of interest.

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