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DIELECTRIC PROPERTIES OF EPOXY NANOCOMPOSITES USING CARBON BLACK AND MULTIWALLED CARBON NANOTUBE AS A FILLER

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ABSTRACT

An attempt is made to study the effect on dielectric properties of Multiwalled Carbon Nanotube (MWCNT) and Carbon Black (CB) reinforced epoxy composites. For that MWCNT & CB (of different weight %) reinforced epoxy composite were prepared by dispersing the filler in resin. Samples were prepared by solution casting process and characterized for their dielectric properties such as dielectric constant (ε') and AC conductivity (σ_{ac}). The main objective is the investigation of the dielectric properties of the prepared samples at different weight % of the two different filler at changed temperatures and frequency. There are two mechanisms of electrical conduction, first the leakage current obtained by the formation of a percolation network in the matrix and the other by tunneling of electrons formed among conductors nearby (tunneling current); here we are getting conduction by the first mechanism. Dielectric constants of the prepared composite increases with increase in temperature and decreases with an increase in frequency from 0.5 kHz to 10 kHz The peak height of the transition temperature decreases with increasing frequency. This study shows that the type of filler can modify considerably the electrical behaviour of epoxy nanocomposites.

Keywords- Dielectric properties, Tunneling current, MWCNT, Carbon Black, Epoxy, Nanocomposites

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I. INTRODUCTION

Carbon Nanotube (CNTs) is in focus now days due to their exceptional electrical, mechanical, and thermal properties. Reinforcement of small amounts of nanotubes in an insulating polymeric matrix found to render the electrical properties of the composite. The electrical properties of the polymer composites may vary from those of an insulating material to conductive composite with filler network depending on the concentration, property of the conducting fillers and dispersion of conducting fillers in polymeric matrix [1]. Recently, carbon nanotubes are being very much investigated by the researchers globally with keen interest for the industrial applications. Since their discovery in 1991, CNTs have attracted enormous attention in research for their properties and their use in wide industrial applications. Single-wall nanotubes (SWNTs) were widely recognized in regard of their predicted properties among all types of CNT's. The remarkable properties of SWNTs are

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limited in usage because of their high cost and low yield of production. Multiwall carbon nanotubes (MWCNTs) are one of the most common and widely used nano filler for enhancing the conductivity of the polymer composites. MWCNT consist of multiple rolled layers (concentric tubes) of graphene shown in fig. 2 with several concentrically aligned tubular graphene sheets, and the typical diameter in the range 8-30 nm [2]. Multiwall carbon nanotubes can be used as reinforcing fillers for different components of composite materials with polymer, metal or ceramic matrices [3] as chemical sensors by using laterally grown MWNT [4, 5] as components of catalytic activity [6, 7] as electromagnetic interference shielding materials [8, 9, 10] for biomedical applications such as selective drug delivery [11, 12]. The most important applications of the CNTs are in the reinforcement of the different types of polymer (thermosetting and thermoplastic) matrix composites due to their high electrical, mechanical and extraordinary thermal conductivity.

The intrinsic potential of carbon nanotubes as reinforcing filler in elastomeric materials is very interesting. Despite a poor dispersion, small filler loading improves substantially the electrical and mechanical behavior of the soft matrix. Bokobza et al [13] showed that, the high electrical conductivity of carbon nanotube-filled composites obtained at a relatively low volume fraction is one of the major attributes of carbon nanotubes since that allows retaining the desired mechanical properties. In polymer composites, dispersion of the used filler as well as interfacial interactions of the matrix and filler has been shown to be essential parameters for enhanced properties. The biggest challenge nowadays is to obtain a homogeneous dispersion of carbon nanotubes in a polymer matrix. This is because of the Vander Waals interactions between individual tubes which often lead to significant aggregation or agglomeration, and causes the reduction of the expected property improvements of the resulting composite [13]. The conduction mechanism in CNT composites has been explained by considering that conductive path formed due to the CNT filler, causing the material to convert from an insulator to a conductor. These conductive paths are formed in the composite when the CNT concentration ϕ increases over a threshold value ϕ_c. The dependency of the conductivity σ on the filler concentration is described by the percolation theory and by a scaling law of the form $\sigma = \sigma_o(\phi - \phi_c)^t$ where ϕ_c is the percolation threshold and t an exponent depending on the system dimensionality [14]. The electrical property of the MWCNT-epoxy composites also depend upon many factors i.e. length and alignment of MWNT, curing of composite [15], dispersion of MWNT in epoxy etc. In recent decades CNTs have been intensively studied as a promising candidate for epoxy resins at low content. If the filler content is not sufficient for making the percolation network then in that case at the onset of percolation, the charge transport follows a thermal fluctuation induced tunneling mechanism, in which the electrons through the thermally induced fluctuating potential barrier formed by a thin insulating polymer layer separating MWCNTs aggregates.

Among the available fillers, carbon black (CB) has been widely used because of its ability to give high electrical conductivity to an insulating polymer at relatively low filler content and low cost. Conductivity of an insulating polymeric material using CB as filler is enhanced by free electron transport through a continuous network in the polymermatrix. Carbon black as filler is used to reduce the tunneling distance and increase the number of tunneling contacts,which have to be overcomeby charge carriers, determining the overall conductivity [16–18].

Recent investigation has shown that epoxy/nanocomposites exhibit some advantages for both mechanical and dielectric properties when compared with pure resin system [19–21]. An enhancement in the electrical

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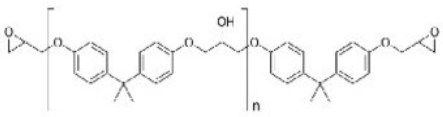
properties of the epoxy nanocomposites can be attributed to the relaxation processes. The relaxation processes correlate to dipolar orientation effects or space charge migration [22,23]. The dielectric effect of the epoxy nanocomposites is due to the charge mobility and interfacial polarization. At lower temperatures, polar side groups enhance the electrical performance of the system. Interfacial polarization is the result of the heterogeneity of the systeme.g., mobile charges assembled at the polymer–filler interface form large dipoles. The volume concentrations of the conductive charges are proved crucial parameter governing the electrical behaviour of the polymer composites [24]. When the filler content is low, the mean distance between charge particles or clusters is large and conductance is limited due to the presence of the dielectric polymer matrix. At a critical volume fraction (or percolating threshold) of the filler, a physical path is formed in a way that the current can flow, percolating the whole system [25]. The effect of CB on the network structure of epoxy composites, like volume fraction of the network, the extent of CB reinforcing, and the interparticle distance between conductive particles, has been investigated in detail for thermally cured and as prepared samples. It is found that the conductivity of an insulating epoxy matrix increases continuously with CB content and is well explained by percolation theory. The temperature dependence of the dielectric response has been analyzed below and at near the epoxy glass transition temperature for various CB concentrations.

The aimof the present study is to obtain new information on the dielectric properties of MWCNT – epoxy and CB–epoxy composite under different conditions of different temperatures and frequencies. In addition,we attempt to give extensive experimental results that may lead to a better understanding of network structure and electrical properties of MWCNT–epoxy and carbon black–epoxy composites for practical applications as heating devices and/or conducting composites.

II. EXPERIMENTAL

2.1 Materials

Epoxy resin contains one or more epoxide groups that serve as cross-linking points when the resin reacts with the hardener to form long chains, the polymerization. The hardener has an impact on the matrix structure and the cross linking ratio and by this way the molecular motions.



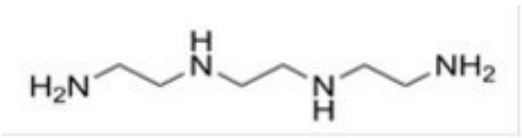


Fig 1(a) Structure of unmodified epoxy pre-polymer resin **Fig 1(b)** structure of a hardener

The thermosetting matrix used in this study was a room temperature cured epoxy resin, provided by M/s Atul Pvt. Ltd. Valsad, India. Figure 1 (a - b) shows the structure of unmodified epoxy pre-polymer resin and structure of a hardener. The density of the resin, cured at room temperature was 1.15 g/cm^3. Industrial grade Multi-wall nanotube (1205YJ) was purchased by Nanostructured & Amorphous Materials, Inc. USA has Purity more than 95%, outside diameter 10-20 nm, inside diameter 5-10 nm, length 10-30 μm, specific surface area 180-230 m^2/g, bulk density 0.04-0.05 g/cm^3.

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Multi-walled carbon nanotubes (MWCNTs), possessing unique mechanical, thermal and electrical properties, as well as excellent processing flexibility and relatively low cost, are ideal nano-fillers for manufacturing low-cost, high-performance nanostructured composite materials. Fig 2 shows the Multi-walled carbon nanotube. The introduction of MWCNTs into polymers can remarkably enhance the mechanical properties, endow the composites with high electric conductivity, and improve abrasion performance and flame resistance of the resulting composites. Normally, all these advantages are considerably affected by the dispersion of MWCNTs in the polymer matrix.

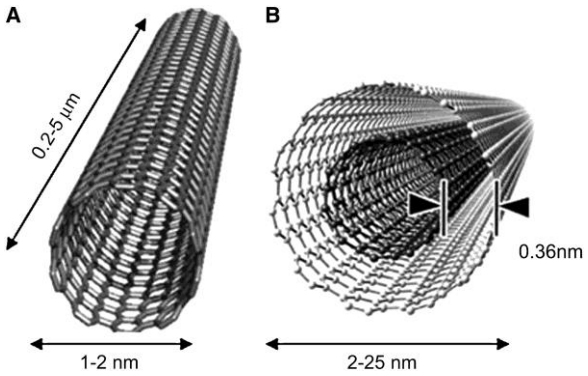

Fig. 2 Multiwall Carbon Nanotube

Carbon black (CB) is an amorphous form of carbon with a structure similar to disordered graphite fig. 3. Various features of carbon black are controlled in production by partially combusting oil or gases. The carbon black used in this study was Ketjenblack EC-600 JD (Supplied by Akzonoble) with a total surface area, BET 1400 m2/g, diameter 36 nm, apparent bulk density 0.12 g/cm3, iodine absorption 1000-1100 mg/g, pore volume DBP 480-510 ml/100g and ash content < 0.1 (as specified). CB is widely used as a reinforcing filler to improve dimensional stability, as a conductive filler to produce conductive polymer composites (CPCs).

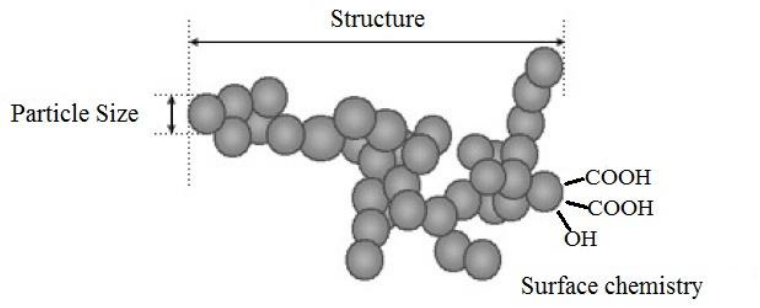

Fig. 3 Carbon Black Surface chemistry

2.2. Composite preparation

Composites filled with different wt% of MWCNT & CB were prepared by the solution casting process. MWCNT & CB both were dispersed individually in the epoxy resin heated at 60 ^{o}C for 30 minutes in an ultrasonic bath sonicator for 30 minutes without any chemical functionalization or surfactant, then reheated the resin/filler mixture at the same temperature and time for reducing the viscosity and kept in a sonicator for another 30 minutes. Increased sonication time may damage the filler. For the homogeneous dispersion of filler in resin, the filler epoxy mixture was stirred by using a lab stirrer at 200 rpm for 60 minutes. The hardener is

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then added in the dispersed filler-epoxy mixture at room temperature. This mixture was cured at 110 ^{o}C for 2 hours. The resin/hardener ratio in this preparation was kept 10:1.

2.3. Test sample preparation

Specimens having a different wt % of MWCNT & CB filled epoxy composites were prepared. The test specimens were cut from the sheets in the sizes of 1 mm thickness and 10 mm diameter. Uniformity of surface was obtained by polishing the specimens. Both sides of the specimens were coated by using air drying type silver conducting paint in such a manner that both the surfaces should not connect electrically with each other. The test specimens were then annealed at 60 ^{o}C for 10 minutes and then kept in between the electrodes of the sample holder for various measurements.

III. CHARACTERIZATION

3.1. Dielectric measurements

The dielectric properties of materials play a key role in the practical performances of integrated circuits. A basic understanding of dielectric properties is therefore needed for engineers and scientists working in semiconductor industries. One important property of dielectric materials is the dielectric constant (permittivity). Dielectric constant (ε') is a measure of the ability of a material to be polarized by an electric field, and closely related to the capacitance (*C*) i.e. the ability to store electric charge. Capacitance (C) and tan δ values were measured by using a Wayne Kerr 6500B Impedance Analyzer in the temperature range from 30^{o}C to 185^{o}C at different frequencies (0.25 kHz to 5 kHz) keeping the heating rate constant at 2 ^{o}C/min. Dielectric constant (ε') of the composite has been calculated by using the following relation

$$\varepsilon' = \frac{C}{Co} \qquad (1)$$

Where C and C_O are the capacitance with and without dielectric, respectively; C_O in pF is given by

$$Co = \frac{(0.08854)A}{d} pf$$

Where A (cm^2) is the area of the electrodes and d (cm) the thickness of the sample.

In dielectric analysis, the sample is placed between two parallel electrodes. By applying a sinusoidal voltage, an alternating electric field is created, due to which polarization is produced in the sample, which oscillates at the same frequency as the electric field, but has a phase angle shift. The phase angle shift is measured by comparing the applied voltage to the measured current, which is separated into capacitive and conductive components [26, 27]. Measurements of capacitance and conductance are used to calculate:

(i) Real part of permittivity (apparent permittivity) ε', which is proportional to the capacitance and measures the alignment of dipoles.

(ii) Imaginary part of permittivity (loss factor) ε", which is proportional to the conductance and represents the energy required to align dipoles and move ions.

(iii) Dissipation factor, tan δ = ε"/ ε'.

AC conductivity ($\sigma_{a.c.}$) was calculated from the relation

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$$\sigma_{a.c.} = \varepsilon_0 \omega \varepsilon' \tan \delta \quad (3)$$

where ε_0 is the permittivity of free space, tan δ the dielectric dissipation factor and ω the angular frequency.

At lower and intermediate frequencies ε' and tan δ values in MWNT reinforced epoxy composites are due to the contributions of orientation, space charge and interfacial polarization. Contribution of orientation polarization decreases at high frequency because the molecules do not have time for orientation which is evidenced by the decrease in ε' and tan δ of composites with frequency.

IV. RESULTS AND DISCUSSIONS

4.1. Dielectric Constant (ε') of EP-MWCNT nanocomposites

Figure 4 (a - f) shows the variation of dielectric constant (ε') with temperature (T) a) pure epoxy (EP-00), (b) 0.5 wt % MWCNT, (c) 1 wt % MWCNT, (d) 1.5 wt % MWCNT, (e) 2 wt % MWCNT and (f) 2.5 wt % MWCNT composite at 0.5, 3, 5, 8 and 105 kHz respectively. It can be ascertained that the dielectric constant (ε') of composites is closely related to the frequency, Figure 4 (a) shows that dielectric constant (ε') increases with increase of temperature from 30 ^{o}C to 180 ^{o}C and decreases with an increase in frequency from 0.5 kHz to 10 kHz. At low frequencies, all the dipole groups in the epoxy molecular chains can orient themselves, resulting in higher dielectric constant. When the frequency of ac voltage increases, the polarization fails to settle itself completely and the values of the dielectric constant of epoxy resin begin to drop approaching at very high frequencies. The peak height of the transition temperature (T_t) decreases with increasing frequency. At lower temperatures ε' values at different frequencies merged. Figure 4 (b) shows that dielectric constant increase with increase in temperature and decreases with the increase of frequency from 0.5 kHz to 10 kHz. It is observed that the ε' increases initially with temperature up to 100 ^{o}C after that it decreases up to 150 ^{o}C and again increases up to 185 ^{o}C Figure 4 (c) shows that the dielectric constant (ε') increases initially with temperature up to 100 ^{o}C and then decreases with temperature until it increased at 150 ^{o}C. The same trend can be seen in Figure 4 (e) with the difference that in this curve the value of the dielectric constant (ε') increases up to 8.5 at 110 ^{o}C for 0. 5 kHz frequency. The reason behind the increase in ε' after 150 ^{o}C may be, when the temperature is lower than a transition temperature (T_t) of the composite the expansion of the polymer matrix will separate the MWCNT fillers which contributes for the conducting path by the electron tunneling before. As the interfaces between the MWCNTs and the polymer matrix will increase by increasing the temperature, the dielectric constant will increase. When the temperature is higher than T_t, the epoxy crystalline phase begins to melt, transforming to the rubbery flow region. This will make it easier for the fillers to connect or transfer the electron tunneling with each other. Therefore interfaces between the MWCNTs and the polymer matrix will decrease hence, the

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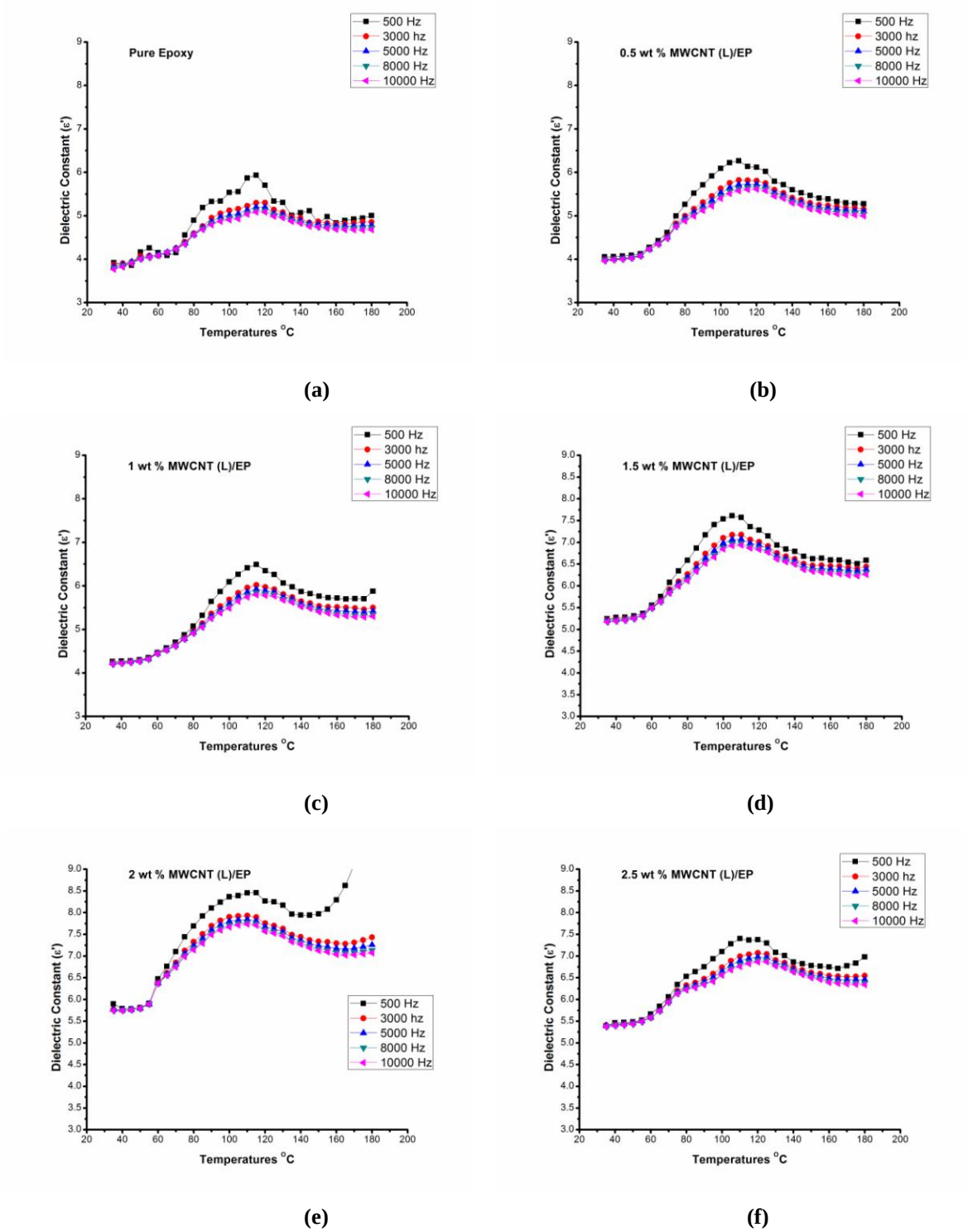

Fig 4(a-f) Shows the variation of dielectric constant (ε') with temperature (T) for (a) pure epoxy, (b) 0.5 wt % MWCNT, (c) 1 wt % MWCNT, (d) 1.5 wt % MWCNT, (e) 2 wt % MWCNT and (f) 2.5 wt % MWCNT composite at 0.5, 3, 5, 8 and 10 kHz respectively

dielectric constant will decrease. On the other hand, the electrons will capture more energy when the temperature increases, and they can overcome the potential barrier easily. So the dielectric constant decreases with increasing temperature. However, the nomadic electrons will get larger energy and agglomerated on the

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interfaces at higher temperatures. The polarization will be enlarged, even exceed the influence of MWCNTs again. The dielectric constant increases again [28].

4.2. Dielectric Constant (ε') of EP-CB nanocomposites

The volume concentration of the fillers in composites has been proved to be a crucial parameter governing the dielectric behaviour of the polymer composites. Figs. 5 (a-f) shows the effect of frequency and temperature on the dielectric constant (ε′) of the prepared CB–EP composites for (a) pure epoxy, (b) 0.5 wt % CB, (c) 1 wt % CB, (d) 1.5 wt % CB, (e) 2 wt % CB and (f) 2.5 wt % CB composite at 0.5, 3, 5, 8 and 10 kHz respectively. When the content of filler in polymer matrix is low the dielectric constant (ε′) of the EP– CB composite changes slightly at all frequencies (Figs. 5). The composites exhibit insulating properties due to the large mean distance between charged particles (carbon black as filler) or clusters and limited conductance due to the presence of the filler in polymer matrix. The conductive CB particles form a finite cluster in the epoxy matrix hence the conductive network paths cannot form in the matrix due to the physical barriers between the gaps. These gaps hinder the flow of charge carriers through the epoxy matrix. A small increase in conductivity of polymer composite may be attributed to the transportation of the small number of charged particles through the polymer matrix without having any continuous conductive path [29-31]. At a critical volume concentration (or percolating threshold) of the filler, a physical path percolating thewhole system is formed within the matrix in away that the current can flow [25]. It is evident from the graphs of Figs. 5 that the volume fraction of the filler affects the dielectric constant (ε′) of the polymer composite considerably.

The peak height at the transition temperature decreases with increasing frequency as shown in Figs. 4 (a-f) and 5 (a-f). At low frequencies the mobile ions accumulate at the interface which gives a high value of dielectric constant as it is a measure of stored charge directly related to charge carriers. Also, all the dipole groups in the epoxy molecular chains can orient themselves at low frequencies again resulting in higher dielectric constant (ε′). With an increase in the frequency of ac voltage, the polarization fails to settle completely and the values of dielectric constant of epoxy resin begin to drop at the higher frequencies. At high frequencies, periodic reversal of the electric field occurs so fast that there is no room for excess ion diffusion in the direction of the field and polarization due to charge accumulation decreases, leading to the decrease in dielectric constant.There are two mechanisms of electrical conduction. One is when the conductors in composites connect with each other to form conductive routes, and the other is that the conductors in composites do not connect with each other, but their distances are so small that the electrons can be transmitted through electron tunnels formed among conductors nearby. Obtained current in first mechanism is named as a leakage current and in second the resultant current is named as a tunneling current. Generally, being higher the leakage current makes more contribution to conductivity than tunneling current. Hence for the leakage current, the content of filler should be high so that a network is formed in the matrix. Thus, the conduction mechanism of the composites is closely related to both the dispersion and the content of conductors. Specifically, when the content of MWCNTs is very small, that is, the content of MWCNTs in the composites is not sufficient to make the connecting networks; hence their conduction mechanisms are due to the tunneling of electron [32].

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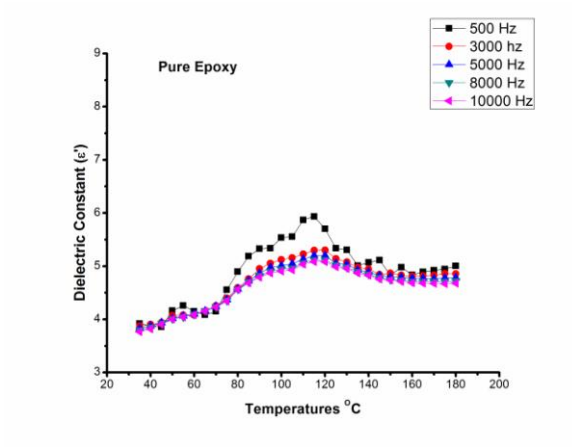

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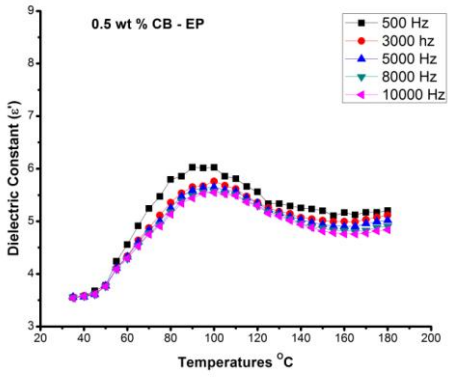

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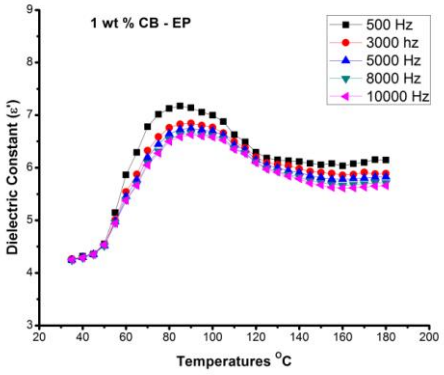

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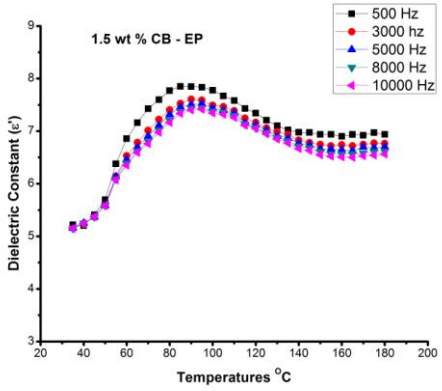

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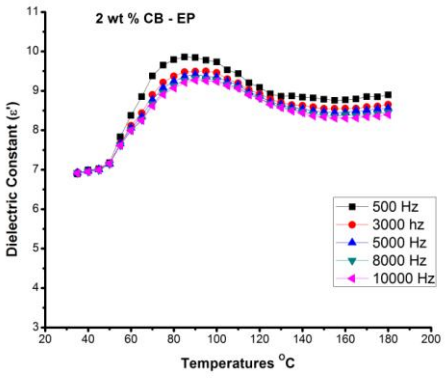

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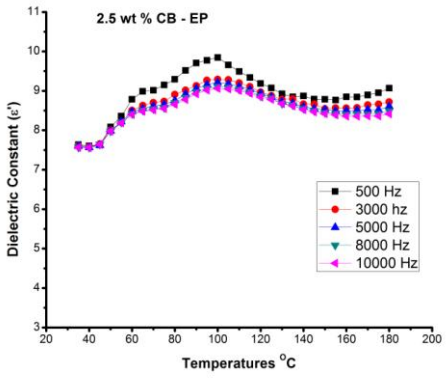

(f)

Fig 5(a-f) Shows the variation of dielectric constant (ε') with temperature (T) for (a) pure epoxy, (b) 0.5 wt % CB, (c) 1 wt % CB, (d) 1.5 wt % CB, (e) 2 wt % CB and (f) 2.5 wt % CB composite at 0.5, 3, 5, 8 and 10 kHz respectively

Analyzing figure 4 and 5 it can be concluded that the dielectric constant (ε') of CB-EP samples are having the higher values as compared to the MWCNT-EP samples at 2 wt % CB in epoxy matrix.

4.3. AC conductivity (σ_{ac})of EP-MWCNT nanocomposites

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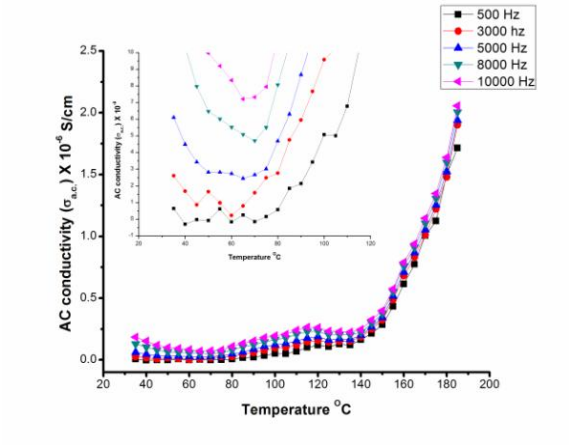
Figure 6 (a-f) show the plots of AC conductivity (σ_{ac}) with Temperature (T) for (a) pure epoxy, (b) 0.5 wt % MWCNT, (c) 1 wt % MWCNT, (d) 1.5 wt % MWCNT, (e) 2 wt % MWCNT and (f) 2.5 wt % MWCNT composite at 0.5, 3, 5, 8 and 10 kHz respectively. These plots show that the AC conductivity increased with increasing temperature and frequency. The inset curves clearly show the variation of the AC conductivity with temperature and frequency up to 160 ^{o}C. It is observed that AC conductivity (σ_{ac}) of all the five samples increases with the increase in temperature and that confirms the negative coefficient of resistance behaviour. The AC conductivity of the sample increased with the increase in wt% of the MWCNT. This behaviour also suggests that the electrical conduction is increasing at the higher temperature which may be again due to the increase in the segmental mobility of the polymer molecules.



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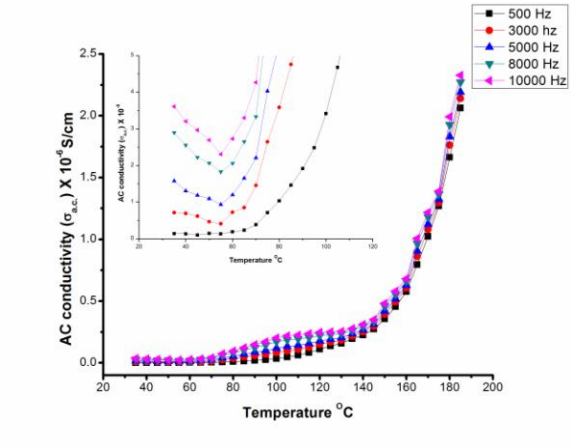

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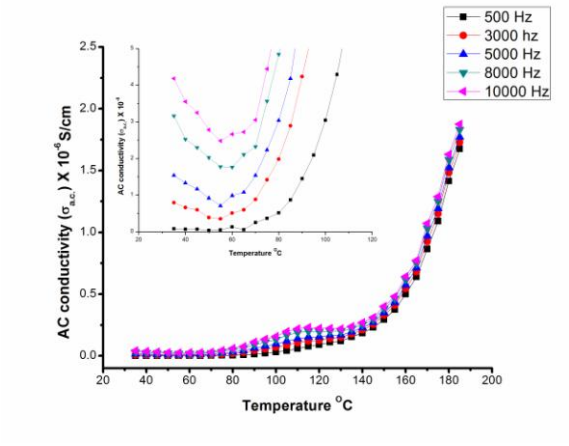

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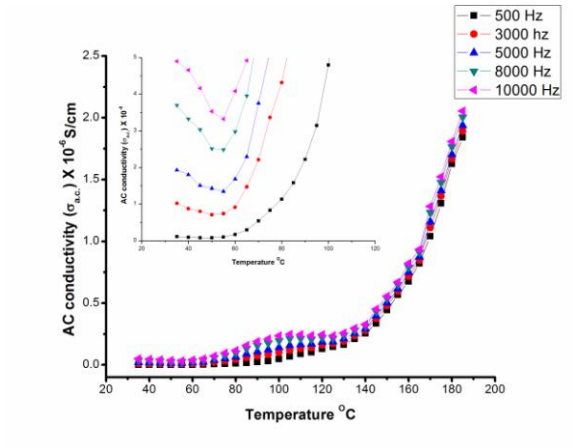

(d)

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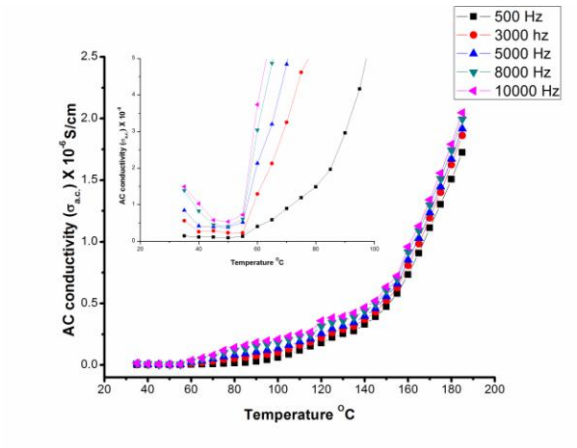
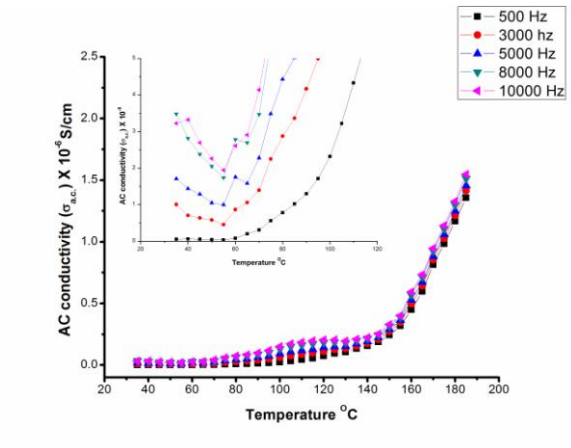


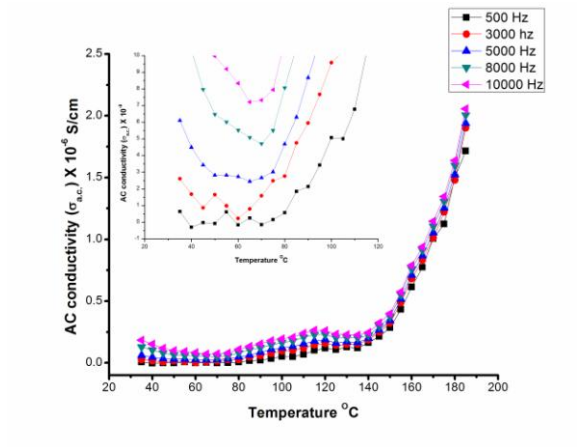


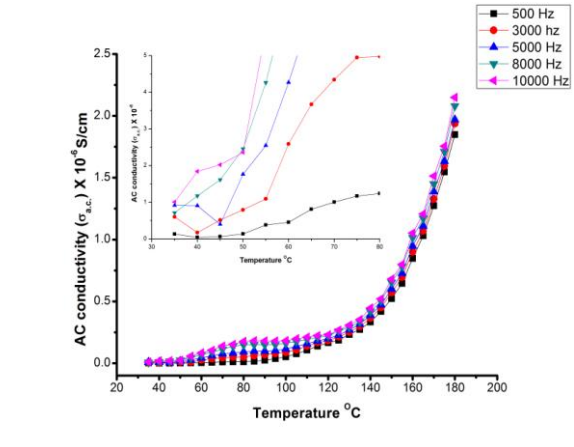
(e) **(f)**

Fig 6(a-f) Shows the plots of AC conductivity (σ_{ac}) with Temperature (T) for (a) pure epoxy, (b) 0.5 wt % MWCNT, (c) 1 wt % MWCNT, (d) 1.5 wt % MWCNT, (e) 2 wt % MWCNT and (f) 2.5 wt % MWCNT composite at 0.5, 3, 5, 8 and 10 kHz respectively

4.4. AC conductivity (σ_{ac})of EP-CB nanocomposites

Figs. 7 (a-f) shows the effect of filler concentration of carbon black on the AC conductivity (σ_{ac}) with different frequencies for the batches of prepared samples. The inset graph of these plots shows that, AC conductivity increases with increasing filler concentration and temperature. The increase in σac is more for the higher filler concentration as compared to the pure epoxy. This increase in σac starts from50 °C for different filler concentrations. Before this temperature the σac value remains almost the same for all the filler concentration samples.





(a) **(b)**

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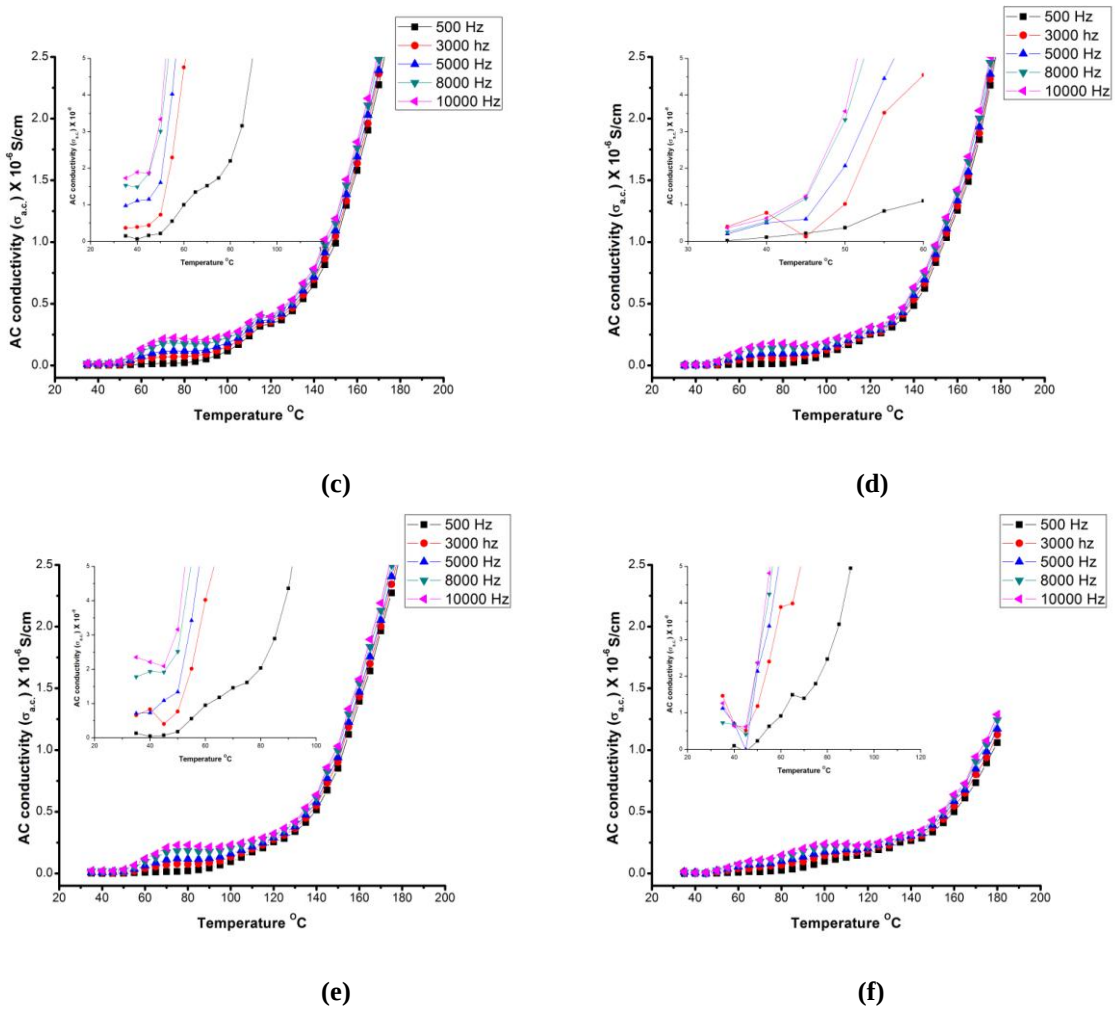

Fig 6(a-f) Shows the plots of AC conductivity (σ_{ac}) with Temperature (T) for (a) pure epoxy, (b) 0.5 wt % CB, (c) 1 wt % CB, (d) 1.5 wt % CB, (e) 2 wt % CB and (f) 2.5 wt % CB composite at 0.5, 3, 5, 8 and 10 kHz respectively

Figs. 5 and 6 show the dependency of the AC conductivity (σ_{ac}) on the frequency for different wt % fillers. The increase in conductivity by increasing the frequency and temperature is a common response for polymeric and semiconductor samples [33]. The reason for this increase in AC conductivity (σ_{ac}) is tremendous increase of the mobility of charge carriers in the polymer composite. It is observed from the inset graph of Figs. 5 and 6 that AC conductivity (σ_{ac}) suddenly increases after 80 °C (approximately) at 0.5 kHz. This is because T_g (glass transition temperature) of epoxy is around 75 °C, and below that temperature the AC conductivity increases gradually but the increase is not appreciable. Above T_g, epoxy comes in amorphous phase, and sudden changes are observed in conductivity. For higher frequencies, (i.e., 5 kHz and 10 kHz) σ_{ac} values are suddenly increasing after 50 °C for both batches of prepared samples.

4.5. Dielectric constant and AC Conductivity with log f

Figure 7(a), 7(b) and 8(a), 8(b) shows the variation of ε' and (σ_{ac}) with log f (frequency) for both batches of prepared samples at 100 ^oC respectively. It was observed that the ε' decreased with increasing frequency and a.c. conductivity increased with increasing frequency. The change of ε' at lower frequency region is higher than that of at high frequency. In AC conductivity an interesting point was observed that the sudden increase seen in the

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value at 1 kHz frequency for EP CNT 0.5 whereas a gradual increase in the value of EP CNT 0.1 and EP CNT 0.3.

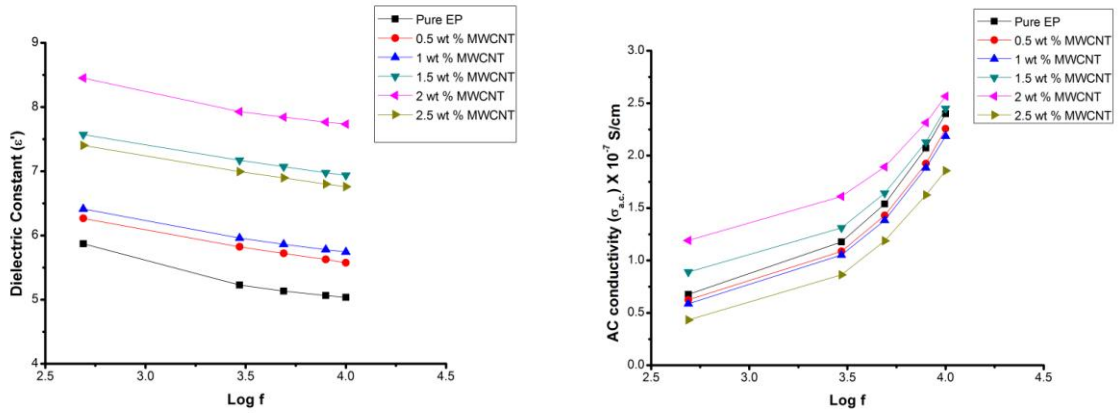

Fig 7(a) & 7(b) Shows the variation of dielectric constant (ε') and AC Conductivity (σ_{ac}) with log f (frequency) for (a) pure epoxy, (b) 0.5 wt % MWCNT, (c) 1 wt % MWCNT, (d) 1.5 wt % MWCNT, (e) 2 wt % MWCNT and (f) 2.5 wt % MWCNT composite at 100 ^{o}C

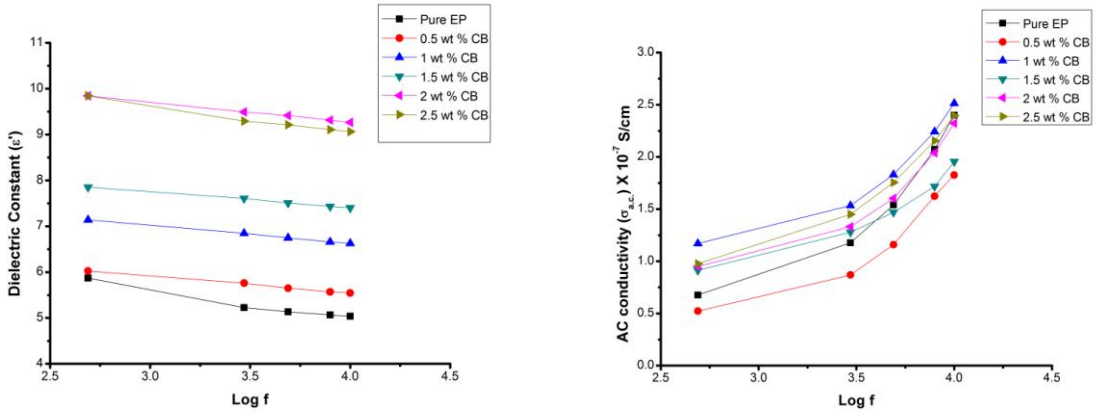

Fig 8(a) & 8(b) Shows the variation of dielectric constant (ε') and AC Conductivity (σ_{ac}) with log f (frequency) for (a) pure epoxy, (b) 0.5 wt % CB, (c) 1 wt % CB, (d) 1.5 wt % CB, (e) 2 wt % CB and (f) 2.5 wt % CB composite at 100 ^{o}C

It is clear from the figure that ε′ values decrease with the increase in the log (f) for all prepared filler concentrations. The ε′ values are approximately the same for 2 and 2.5 wt % CB-EP composite. The change of ε′ at lower frequency region is higher than that of at high frequency. The atomic and electronic polarizations are instantaneous polarization components, the effect of which is seen only at high frequencies. The dipole or orientation polarization occurs due to the presence of polar groups in the material. The interfacial polarization arises due to heterogeneity, which is higher at lower frequency. Hence, the higher values of ε′ at low frequency can be explained in terms of interfacial polarization. As observed the σ_{ac} value increases with increase in log (f).With all filler concentrations, the increase of frequency increases AC conductivity, which is due to increase in the hopping of conducting electrons present in filler. At higher frequencies this hopping frequency could not match the applied field frequency.

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V. CONCLUSIONS

The small quantity addition of filler can modify considerably the electrical behaviour of a epoxy nanocomposite. The conduction mechanism of the polymer nanocomposites is closely related to both the dispersion of the filler as well as the contents of the filler. At low weight % of MWCNT in epoxy composites, the conduction was due to the charge transport which follows through a thermal fluctuation induced tunneling mechanism in place of percolation network formation. The type of filler plays an important role in the conduction mechanism. Here in this study the carbon black as a filler enhances the dielectric constant (ε') of the prepared sample more as compared to the Multiwalled carbon nanotube, which can effect the industrial production of the epoxy nanocomposites. MWCNT is costly as compared to CB, hence using CB as filler we are getting the higher dielectric constant (ε'). At low weight % of MWCNT in epoxy composites, the conduction was due to the charge transport which follows through a thermal fluctuation induced tunneling mechanism in place of percolation network formation.

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REFERENCES

[1] F. Lux, (1993) Models proposed to explain the electrical conductivity of mixtures made of conductive and insulating materials. J. Mater. Sci. 28: 2: pp 285-301.

[2] Ilya Mazov, Vladimir Kuznetsov, Anatoly Romanenko and Valentin Suslyaev. Properties of MWNT-Containing Polymer Composite Materials Depending on Their Structure Chapter 3 2012 InTech http://dx.doi.org/10.5772/48245

[3] J. Wang, H. Kou, X. Liu, Y. Pan, J. Guo (2007) Reinforcement of mullite matrix with multiwalled Carbon nanotubes. *Ceram. Int.* 33: 719–722.

[4] Y.-T. Jang, S.-I. Moon, J.-H. Ahn, Y.-H. Lee, B.-K. Ju (2004) A simple approach in fabricating chemical sensor using laterally grown multi-walled carbon nanotubes. *Sens. Actuators, B* 99: 118–122.

[5] N. Sinha, J. Ma, J.T.W. Yeow (2006) Carbon Nanotube-Based Sensors. *J. Nanosci. Nanotechnol.* 6: 573–590.

[6] N. Maksimova, G. Mestl, R. Schlogl (2001) Catalytic activity of carbon nanotubes and other carbon materials for oxidative dehydrogenation of ethyl benzene to styrene. *Stud. Surf. Sci. Catal.*, 133: 383–389.

[7] M.S. Saha, R. Li, X. Sun (2008) High loading and monodispersed Pt nanoparticles on multiwalled carbon nanotubes for high performance proton exchange membrane fuel cells. *J. Power Sources* 177: 314–322.

[8] M.H. Al-Saleh, U. Sundararaj (2009) Electromagnetic interference shielding mechanisms of CNT/polymer composites. *Carbon* 47: 1738-1746.

[9] I.W. Nam, H.K. Lee, J.H. Jang (2011) Electromagnetic interference shielding/absorbing characteristics of CNT-embedded epoxy composites. *Composites: Part A* 42: 1110–1118.

IJARSE
ISSN (O) 2319 - 8354
ISSN (P) 2319 - 8346

[10] Ki-Yeon Park, Sang-Eui Lee, Chun-Gon Kim, Jae-Hung Han (2007) Application of MWNT-added glass fabric/epoxy composites to electromagnetic wave shielding enclosures. *Composite Structures* 81: 401–406

[11] T.A. Hilder, J.M. Hill (2008) Carbon nanotubes as drug delivery nanocapsules. *Current Applied Physics* 8: 258 - 261

[12] C. Tripisciano, K. Kraemer, A. Taylor, E. Borowiak-Palen (2009) Single-wall carbon nanotubes based anticancer drug delivery system. *Chem. Phys. Lett.* 478: 200–205

[13] Liliane Bokobza (2007) Multiwall carbon nanotube elastomeric composites: A review. *Polymer* 48: 4907-4920

[14] L. Guadagno et. al. (2011) Effect of functionalization on the thermo-mechanical and electrical behavior of multi-wall carbon nanotube/epoxy composites. CARBON 49: 1919–1930.

[15] Manindra Trihotri, U.K. Dwivedi, Fozia Haque Khan, M.M.Malik, M.S. Qureshi (2015) Effect of curing on activation energy and dielectric properties of carbon black–epoxy composites at different temperatures. *J. Non-Cryst. Solids* 421:1–13.

[16] W.S. Chin, D.G. Lee, Dielectric characteristics of E-glass–polyester composite containing conductive carbon black powder, J. Compos. Mater. 41 (2007) 403–417.

[17] O.G. Abdullah, G.M. Jamal, D.A. Tahir, et al., Electrical characterization of polyester reinforced by carbon black particles, Int. J. Appl. Phys. Math. 1 (2011) 101–105.

[18] Z.M. Elimat, AC-impedance and dielectric properties of hybrid polymer composites, J. Compos. Mater. (2013) 1–13, http://dx.doi.org/10.1177/0021998313514256.

[19] J. Katayama, Y. Ohki, N. Fuse, M. Kozako, T. Tanaka, Effects of nanofiller materials on the dielectric properties of epoxy nanocomposites, IEEE Trans. Dielectr. Electr. Insul. 20 (1) (2013)http://dx.doi.org/10.1109/TDEI.2013.6451354.

[20] Q. Wang, G. Chen, Effect of nanofillers on the dielectric properties of epoxy nanocomposites, Adv. Mater. Res. 1 (1) (2012) 93–107.

[21] M.E. Achour, A. Mdarhri, F. Carmona, F. Lahjomri, A. Oueriagli, Dielectric properties of carbon black–epoxy resin composites studied with impedance spectroscopy, Spectrosc. Lett. 41 (2008) 81–86, http://dx.doi.org/10.1080/00387010801943848.

[22] R. Strumpler, J. Glatz-Reichenbach, Conducting polymer composites, J. Electroceram. 3 (4) (1999) 329–346.

[23] J.P. Runt, S. Fitzgerald (Eds.),Dielectric spectroscopy of polymeric materials, J. Am. Chem. Soc., 119 (49), 1997http://dx.doi.org/10.1021/ja975608v (12029–12029).

[24] N.L. Singh, S. Shah, A. Qureshi, A. Tripathi, F. Singh, D.K. Avasthi, P.M. Raole, Effect of ion beam irradiation on metal particle doped polymer composites, Bull. Mater. Sci. 34 (1) (2011) 81–88, http://dx.doi.org/10.1007/s12034-011-0040-5.

[25] M. El Hasnaoui, J. Belattar, M.E. Achour, L.C. Costa, F. Lahjomri, Electrical transport properties of CB/epoxy polymer composites, Optoelectron. Adv. Mater. Rapid Commun. 6 (5–6) (2012) 610–661.

[26] I. Ben Amora, H. Rekik, H. Kaddami, M. Raihane, M. Arous, A. Kallel (2009) Studies of dielectric relaxation in natural fiber–polymer composites. *J. Electrost.* 67: 717–722.

IJARSE
ISSN (O) 2319 - 8354
ISSN (P) 2319 - 8346

[27] Manindra Trihotri, Deepak Jain, U. K. Dwivedi, Fozia Haque Khan, M. M. Malik, M. S. Qureshi (2013) Effect of silver coating on electrical properties of sisal fibre-epoxy composites. Polym. Bull. (2013) 70:3501–3517.DOI 10.1007/s00289-013-1036-7.

[28] Q. Li1, Q. Z. Xue, X. L. Gao, Q. B. Zheng. (2009) Temperature dependence of the electrical properties of the carbon nanotube/polymer composites. eXPRESS Polymer Letters Vol.3, No.12-769–77.

[29] M. El Hasnaoui, M.P.F. Graça, M.E. Achour, L.C. Costa, F. Lahjomri, A. Outzourhit, A. Oueriagli, Electrical properties of carbon black–copolymer composites above and below the melting temperature, J. Mater. Environ. Sci. 2 (1) (2011) 1–6.

[30] F. El-Tantawy, K. Kamada, H. Ohnabe, In situ network structure, electrical and thermal properties of conductive epoxy resin–carbon black composites for electrical heater applications, Mater. Lett. 56 (2002) 112–126.

[31] Manindra Trihotri, U.K. Dwivedi, Fozia Haque Khan, M.M.Malik, M.S. Qureshi (2015) Effect of curing on activation energy and dielectric properties of carbon black–epoxy composites at different temperatures. J. Non-Cryst. Solids 421:1–13.

[32] Jianfei Chang, Guozheng Liang, Aijuan Gu, Shiduan Cai, Li Yuan (2012) The production of carbon nanotube/epoxy composites with a very high dielectric constant and low dielectric loss by microwave curing. Carbon 50: 689 –698.

[33] P. Dutta, S. Biswas, M. Ghosh, S.K. De, S. Chatterjee, The dc and ac conductivity of polyaniline and polyalcohol blends, Synth. Met. 122 (2000) 455–461.