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Dielectric Properties and AC conductivity of (Epoxy / Ion exchange) blend

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Abstract

The dielectric behavior of blend materials epoxy resin- Polyvinyl benzyl dimethyl ethanol ammonium chloride (PBDEAC) (Ion exchange) was analysed as a function of Ion exchange weight content; temperature and frequency. Blends were prepared by mixing the components and pouring them into suitable moulds. The dielectric parameters have been measured using parallel plate capacitor method in the frequency range from 120 Hz to 2 MHz and in the temperature range from 30 °C to 110°C. Variations of real (ε′) and imaginary (ε") parts of dielectric constants and loss tangent of material with frequency and temperature have been studied. The experimental results showed that (ε′) and (ε") increased with the addition of Ion exchange filler content. The value of (ε′) decreased with increasing frequency, which indicates that the major contribution to the polarization comes from orientation polarization. Dielectric loss peaks were also observed in the composite materials at high temperature. The value of (ε′) increased with increasing temperature, and is due to greater freedom of movement of the dipole molecular chains within the polymer at high temperature. Ac. conductivity and impedance of the composites behaviours as function of frequency and temperature have also been investigated.

Keywords: Dielectric properties, epoxy, Ion exchange, ac. conductivity

1. Introduction

Development of electronic devices working at high operating frequencies, such as fast computers, cellular phones, etc., requires new high dielectric permittivity materials that combine good dielectric properties with both mechanical strength and ease of processing. In particular, the high dielectric permittivity materials are required for making embedded capacitors for integrated electronic devices (Abdulmajeed *et al.*2012 & Haines *et al.*2010).

Dielectric behavior depends on the polarization that is induced in a material due to the effect of an external electrical stress. Dielectric spectroscopy can provide useful information on the electrical properties of the specimens. Moreover, this technique can be used as analytical tool whereby the dielectric data is related to other properties such as the polymer molecule structure or morphology (Haines *et al.*2010), or its degradation and ageing.

Polymer blend is one of the major subjects in polymer industry. The purpose of alloying or blending one polymer system with another polymer is to gain synergistic improvement in properties at both the micro and macro levels. Polymeric insulating materials such as Epoxy and polyethylene are widely used in manufacturing weather shed of outdoor insulators and cable terminators (Muhammad *et al.*2005, Bielinski *et al.*2001, Sadek *et al.*2003, Irinislimane *et al.* 2007).

Epoxy resins are widely used as suitable matrices, besides their other applications in modern technology, because they offer versatility, low shrinking, chemical resistance, relativity low dielectric constant and outstanding adhesion (Psarrasa *et al.* 2003). Epoxy blends are very popular insulators in high voltage engineering, such as epoxy mica composites which are used for the insulation of power transformers, station and line posts, insulation of coils in rotating machines, etc. (Gonon *et al.* 2005). The Predication of the dielectric permittivity of blend materials is very important in many relevant technological applications (Ramajo *et al.* 2008). Ion exchange materials are insoluble substances containing loosely held ions which are able to be exchanged with other ions. These exchanges take place without any physical alteration to the ion exchange material (Pravin *et al.* 2012).

The relationship between capacitance, C and the dielectric constant (ε′) of the ferrite sample calculated using the formula (Tatarog *et al.* 2005, Hussein *et al.* 2012 , Yang *et al.*2007& Hussein *et al.*2010)

$$C = \varepsilon_o\, \varepsilon'\, A / D \quad \ldots\ldots\ldots (1)$$

where ε_o (permittivity of vacuum) is the dielectric constant of the free space(8.854×10^{-12}F/m), C is the capacitance in farads, the area “A” is in square metres and it represents the distance between electrodes and “D” the thickness of the insulator is in meters. The complex dielectric constant (ε") of the ferrite sample is given by

$$\varepsilon'' = \varepsilon' \tan\delta \quad \ldots\ldots\ldots (2)$$

$$D = \tan\delta \quad \ldots\ldots\ldots (3)$$

Where D is the Dissipation Factor and tan δ is loss tangent

The ac. conductivity σ_{ac} for an ionic electrolyte material is calculated from the relation

$$\sigma_{a.c} = \varepsilon_o . \varepsilon'' . \omega \ldots\ldots\ldots (4)$$

Where D is the ω angular frequency**,** equation (4) is often called " The ac universality law **".**

2. Experiment

2.1 Materials

The epoxy studied is a traditional grad in the local market of Basrah city, which is produced by United Chemical Company Ltd. (UNICHEM, JORDEN) type diglycidylether of bisphenol-A (DGEBA-368WG) Mw = 624 gm / mole, density =1.27 gm /cm^3, the curing agent is a triethylene tetra amine (TETA) supplied by the same company .

Polyvinyl benzyl dimethyl ethanol ammonium chloride (PBDEAC) filler (Ion exchange-Purolite A510) was supplied by (PUROLITE COMPANY LIMITED- China), diameter grain size =45 µm.

2.2 Sample preparation

Ion exchange (PBDEAC) were added to the epoxy resin (as received) at different weight fraction and then mixed at about 70^{0}C for 5 minutes, then curing agent triethylene tetra amine (TETA), the hardener was added and mixed for 5 minutes also. The mixture, then cast on clean Al substrates.

The initial curing was carried at room temperature for 24 hours, followed by post curring at 120^{o}C for 2 hours, Circular aluminum electrodes in diameter (6mm) were vacuum deposited on the upper side of the blend sample, and finally thermosetting sheets of epoxy blends were made.

2.3 Characterization and measurements:

The samples capacitance and the loss tangent (tanδ) of blends were measured by Digital RCL bridge type (MEGGER B131), at the frequencies 120 Hz and 1kHz. At the frequencies in the range (120Hz -2MHz), RCL bridge type (METRAPOINT-RLC2 and ME 1634 FUNCTION GENERATOR) were used to measure the capacitance of the samples. The impedance Z at different frequencies up to 500 kHz was measured by RCL bridge type (hp A 4800).

3. Results and Discussion

The variation of permittivity (dielectric constant) of the pure epoxy resin and epoxy composites as a function of (PBDEAC) filler weight fraction content is shown in figure (1) for two different frequencies (120Hz and 1kHz), respectively. The addition of filler Ion exchange to the epoxy resin increases the permittivity of the prepared composite. The increase in ε′ with both the increase in filler content and lower frequency is attributed to Maxwell-Wagner Sillars (MWS)/ or interfacial effect appears in heterophase systems (Lu *et al.* 2006 & Van Beek *et al.* 1960).

Temperature dependence of dielectric permittivity (ε') in the range (30 – 110)^{o}C at constant electric field frequency at 1kHz of the of (PBDEAC/ Epoxy) blends is shown in figure (2). It can be seen that, permittivity increases as the temperature increases. This is due to the increase in segmental mobility of the dipoles in the matrix (greater freedom of movement of dipole molecular chain of polymer at high temperature), and change in the dielectric response of the filler particles with temperature (Kim *et al.* 1998).

Figure (3) shows the dependence dielectric permittivity (ε') of (PBDEAC/ Epoxy) blends on the frequency in the range (120Hz - 2MHz) for different weight fractions of PBDEAC at room temperature. It is obvious that, the dielectric permittivity decreases with increasing frequency for all of the filler contents. It is believed that a decrease in the dipolar polarization of the matrix and the accumulation of charges at the interface between filler particles and polymers result in a large scale field distortion (Salih *et al.* 2010). After about 100 kHz, the changes become smaller.

Figure (4) shows the variation of dielectric loss (ε'') as a function of filler weight fraction of filler and temperature, at a constant frequency (1 kHz) applied field for (PBDEAC– epoxy) blends. The dielectric loss increases in general as the filler contents and temperature increase. The increase in dielectric loss with the increase of filler contents is related to the interfacial polarization. The increased loss of the blends at higher temperature is related to dipole molecules tendency or parts of dipole molecules that rotate along the electric field overcoming the forces of internal friction of either the intermolecular interaction or the molecular friction, resulting in the expenditure of a part of electric energy and its conversion into thermal energy (Kim et al. 1997 & Shriprakash et al. 2007).

Figure (5) shows the variation of dielectric loss (ε'') with frequency in the range (120Hz - 2MHz) at room temperature for all samples. These kind of frequency dependency is called dielectric relaxation, being characterized by a relaxation time τ or relaxation frequency f_o corresponding to Δε/2. This relaxations shift to lower frequencies with the increasing of PBDEAC content. It can be observed that, the values of dielectric loss increase gradually with increasing the frequency to reach the highest value (maximum value). The increases in dielectric loss may be related to a.c conductivity (σ_{ac}) which depends on the number of charge carriers, relaxation time and frequency domain when the temperature is constant, after that the dielectric loss begins to decrease gradually. This is due to the relaxation processes which were influenced by the interfacial polarization effect (Suzhu *et al.* 2000 & Lipatov *et al.* 1976).

The plots of relaxation time (τ) against the PBDEAC content are shown in figure (6). It is obvious that, the relaxation time increases with increasing filler content. The increases in (τ) with the increase in filler content may be related to the increase in a.c conductivity and is attributed to Maxwell-Wagner Sillars (MWS)/ or interfacial polarization that appears in complex system exhibiting electrical heterogeneity, due to the accumulation of charges at the interfacial of the system (Chaudhari *et al.* 2012). The relation between the dielectric loss ε'' as a function of the dielectric permittivity ε' for (PBDEAC/ Epoxy) blends at room temperature is shown in figure (7). From the Cole-Cole plot, the generalized relaxation time, the parameter relating to the distribution of relaxation times, is determined.

The effect of adding (ion exchange) filler to the epoxy resin on ac conductivity as a function of temperature is shown in figure (8). It can be seen that increase in a.c conductivity with PBDEAC content and temperature is contributed by free charges available in the blend system. It was also found that, at higher temperature, high mobility of free charges makes them more frequency independent conductivity (Saq'an 2004).

The dependence of the calculates a.c conductivity on frequency range (120Hz - 2MHz) for different weight fraction of PBDEAC at room temperature is shown in figure (9) It increased when the filler content increased for all frequency ranges studied. This increase in AC-conductivity is expected, since at higher applied field frequency more ions and impurities are moved. The observed enhancement in the a.c conductivity is attributed to ionic interactions and impurity motion taking in the bulk of electrolyte polymer composites (Bhowmik *et al.* 2010).

Figure (10) Shows the impedance Z (real component) temperature dependence for pure epoxy and epoxy/ PBDEAC blends (different wt%). There is an obvious decrease in Z, with the increasing of filler content due to the increased interfacial polarization, with the rise in temperature.

Figure (11) Shows the variation of impedance Z with frequency for PBDEAC filled epoxy composites. It was noticed that the phase angle was always negative, indicating that the blends were capacitive and could be represented by parallel RC net- works (lumped circuit) connected in series. Impedance values decrease with increasing frequency and PBDEAC concentration. The observed decrease in impedance with PBDEAC content is due to the protonic migration transporting and impurities existing in the PBDEAC filler. This motion leads to higher electrical conduction in the filled blends (Kambale *et al.* 2009). As can be seen, there is an exponential decrease in the impedance with the increase in frequency for all filler weight fractions, and the decrease is greater for high filler contents composites (Krupka 2006).

4. Conclusions

Frequency and temperature dependence of dielectric constant (ε') and dielectric loss factor (ε'') in Epoxy and PBDEAC blends has been investigated in the frequency range (120Hz - 2MHz) and the temperature range (30 – 110)^{o}C. The experimental results indicate that (ε') and (ε'') increased with the addition of PBDEAC in Epoxy. at the lower frequency, the permittivity decreases with increasing frequency, but after about 100 KHz, the changes can be ignored ,and the increase in (ε') with the increase of temperature is due to greater freedom of movement of dipole molecular chain of polymer at high temperature. Dielectric loss peaks in the composite materials at high temperature are due to the glass transition temperature of Epoxy. The ac conductivity and impedance were found to be both frequency and temperature dependent.

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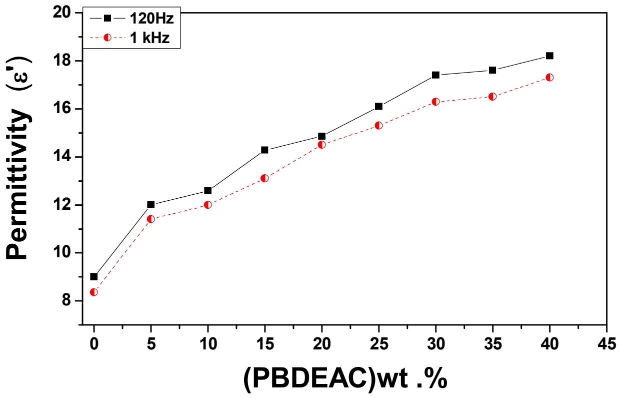

Fig1: The variation of permittivity with two different frequencies (120Hz, 1kHz).

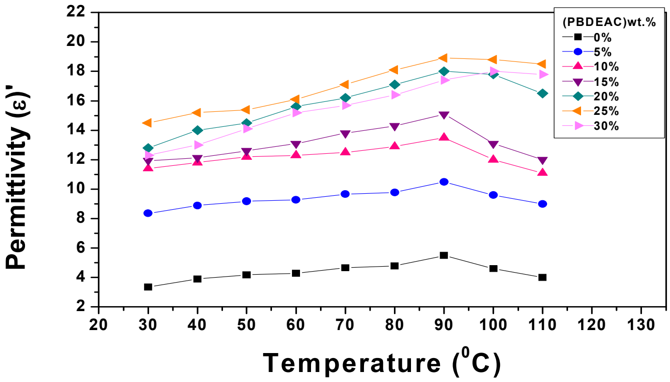

Fig2: PBDEAC-epoxy blends permittivity as a function of filler content and temperature.

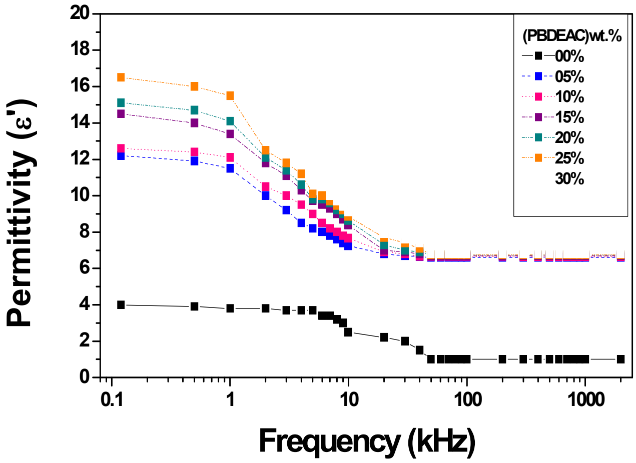

Fig3: The permittivity as a function of filler content and frequency for PBDEAC-epoxy blends.

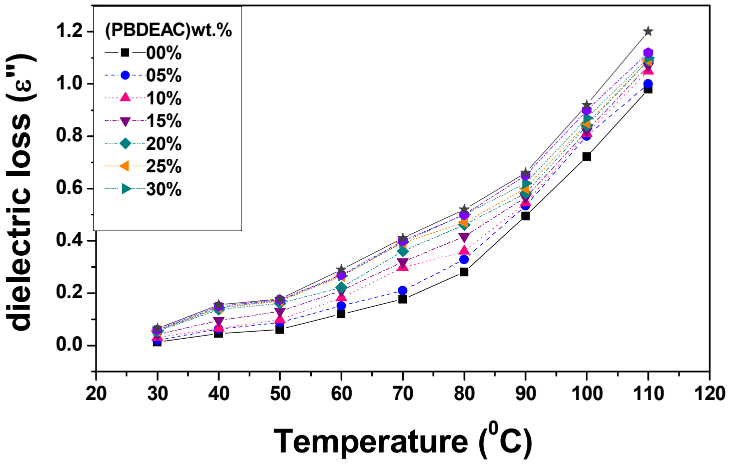

Fig4: The dielectric loss as a function of filler content and temperature for PBDEAC-epoxy blends.

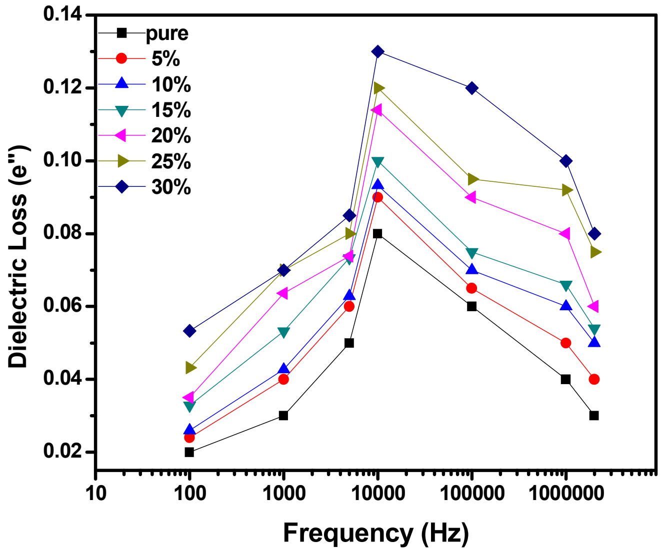

Fig5: The dielectric loss as a function of filler content and frequency for PBDEAC-epoxy blends.

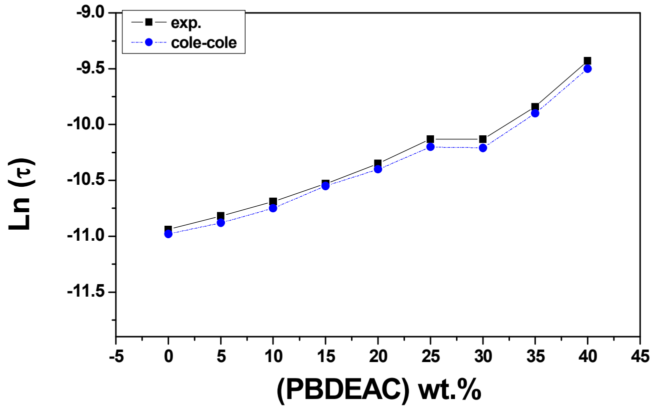

Fig6: The relaxation time as a function of filler content for PBDEAC-epoxy blend .

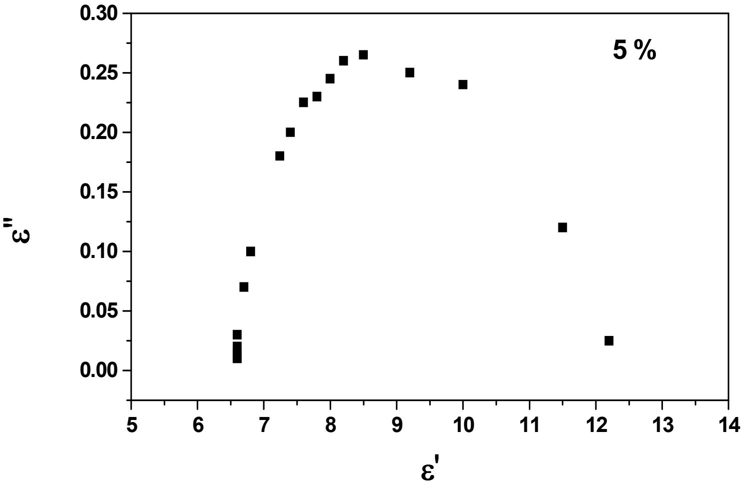

Fig7: The Cole-Cole plot for PBDEAC-epoxy blends.

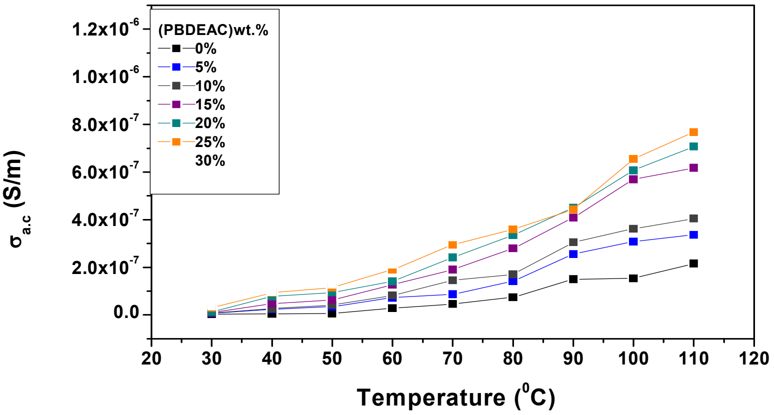

Fig8: The σ_{ac} as a function of filler content and temperature for PBDEAC-epoxy blends.

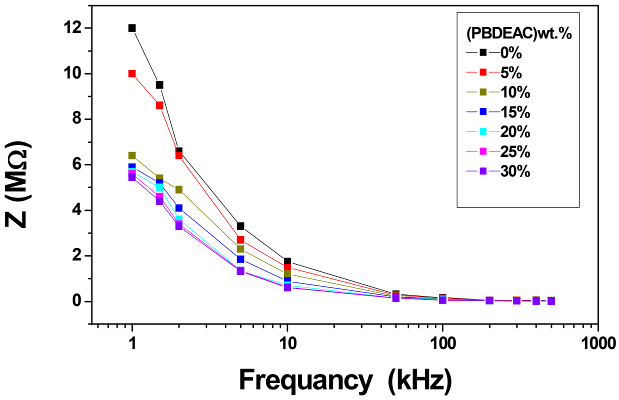

Fig9: The impedance of PBDEAC-epoxy blends as a function of filler content and frequency.

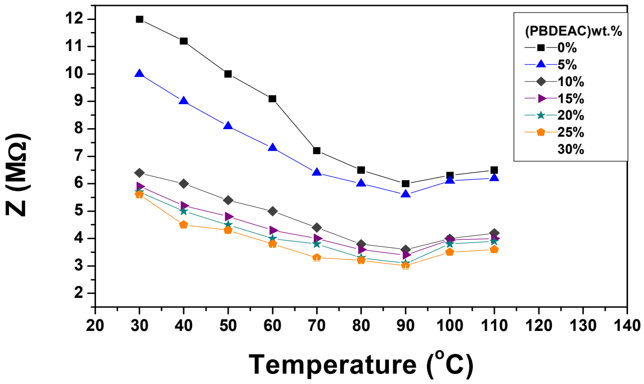

Fig10: The impedance of PBDEAC-epoxy blends as a function filler content and temperature.

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