

Study of A.C Electrical Properties of Aluminum–Epoxy Composites

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Abstract

This work studies the electrical properties for pure epoxy (EP), and epoxy with (20%,30%,40% and 50%)aluminum powder composites. The electrical conductivity [$\sigma(w)a.c$] for these materials have been measured over the frequency range ($10^2 - 10^6$)Hz at room temperature. It is found the A.C conductivity increased when increased the concentration of aluminum.

It is found for all samples, the curves for real part of dielectric constant (ϵ) and dissipation factor ($\tan\delta$). The dielectric constant increased smoothly with an increase in the concentration of aluminum it was attributed to interfacial polarization and decrease with increased frequency. In general, dissipation factor values for composites with higher concentrations of aluminum were greater than those with lower volume content of aluminum. Also, the dissipation factor showed an increase with a decrease in frequency. It was found that a.c electrical conductivity increases with increasing frequency according to the relation $S(w)=Aw^s$ has been observed , with values of exponent (s) less than unity for all samples and decrease with the increase of frequency and lie between (0.3 to 0.6) unit . Also, it was found that the correlated barrier hopping (CBH) is the dominant conduction mechanism.

Keywords: A.C. conductivity, dielectric constant (ϵ), dissipation factor ($\tan\delta$), and exponent (s) factor.

Introduction

Studies of the dielectric properties of polymers have increased importance because it provide an understanding to movement of molecular chains and its applications in electrical and electronic engineering [1].

Epoxy resins are highly cross linked amorphous polymers used for insulation in electric transformers, switchgear, rotating machines, etc. The sensitivity of epoxy composites to humidity is a serious matter of concern because absorption of water may cause significant and possibly irreversible changes to the material [2]. Polymers have a very low concentration of free charge carriers, thus are non conductive, because of its transparency to electromagnetic radiation, they are not suitable for used as an enclosures for electronic equipment because they cannot shield it from outside radiation. Also they cannot prevent the escape of radiation from the component. Polymers cannot provide protection against electrostatic discharge in handling sensitive electronic devices. This drawback has led to the development of electrically conductive polymers such as inherently conductive polyaniline or polymers filled with conductive particles. Beyond a

critical concentration of filler polymer becomes conductive [3]. This formation of a network permits the movement of charge carriers of the fillers through the matrix and so the composites achieve a certain degree of electrical conductivity .Several fillers can be added to the insulating polymeric matrix in order to achieve different conductivity ranges. Filled polymers required for a variety of industrial applications. Graphite epoxy composites can have potential use in the area of thermoelectric power generation. Wu and Tung (1995) reported [4] dielectric properties of pure epoxy resin in the temperature range (-50°C to 70°C) and found relaxation peak in the low temperature range of (-30°C to -20°C) due to the motion of dieter segments introduced in the pure epoxy cross linking network by the reaction of acid anhydride. They studied the relaxation response in a very limited temperature range.

The dielectric study is performed on room temperature and several frequencies this is particularly interesting because of the nature of polymeric systems which have long chain and high molecular weight. It well known that most of the dielectric properties such as

(dielectric constant , dissipation factor , and elastic dispersion) compliance in polymeric materials are dispersive even at low frequencies , this behavior reflecting relatively high activation energies for the motion of molecule unit and chain segment, or dispersive at high frequencies which reflecting relatively low activation energies for the motion of molecule unit and ions .Fig. (1).Illustrate some of the charge configurations and their response (polarization) under the influence of an external field. Because almost material systems are made up of charges (an exception being neutron stars!), it is useful to characterize materials by their dielectric constant. A schematic representation of the real part of the dielectric constant is shown in Fig. (1). At high frequencies ($>10^{14}$ Hz), the contribution comes solely from electronic polarization, implying that only free electrons, as in metals, can respond to the electric field. That is why metals are such good optical reflectors! Even the various thermal and mechanical properties, such as thermal expansion, bulk modulus, thermal conductivity, specific heat, and refractive index, are related to the complex dielectric constant, because they depend on the arrangement and mutual interaction of charges in the material. Thus, the study of dielectrics is fundamental in nature and offers a unified understanding of many other disciplines in materials science [4, 5].

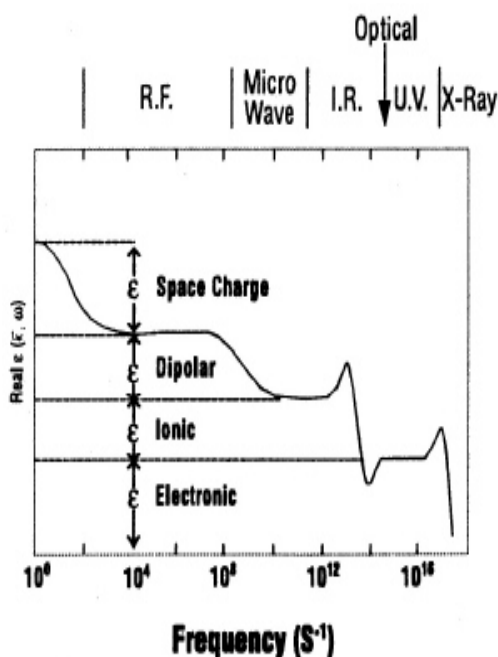


Fig. (1) Contributions to the frequency-dependent dielectric constant from the different charge configurations.

A.C Conductivity Measurements

A.C conductivity is different from the D.C conductivity, where the frequency of electric field is constant during d.c conductivity but during a.c conductivity, frequency of the electric field will be variable. The effect of the two variable parameters, frequency, (ω) and temperature, (**T**), have been studied. A common feature for a.c measurements is that a.c electrical conductivity $\sigma_{a.c}(\omega)$ increased when the frequency is increased, according to the equation [6]:

$$\sigma_{a.c}(\omega) = \sigma_{total}(\omega) - \sigma_{d.c} \dots\dots\dots (1)$$

where ω is the angular frequency ($\omega = 2\pi f$), $\sigma_{total}(\omega)$ is the measured total electrical conductivity, $\sigma_{d.c}$ is the d.c conductivity which depends strongly on temperature and dominate at low frequencies and high temperature , while $\sigma_{a.c}$ is the a.c conductivity which is weaker temperature dependence than $\sigma_{d.c}$ and dominate at high frequency and low temperature then , the empirical relation for the frequency dependence a.c conductivity is given by[5,6] :

$$\sigma_{a.c}(\omega) = A_1 \omega^s \dots\dots\dots (2)$$

where , A_1 is constant ,and(**s**) is a function of temperature and is determined from the slope of a plot $\ln \sigma_{a.c}(\omega)$ versus $\ln \omega$, then (**s**) is not constant but decrease with the increasing temperature usually $0 << 1$ and approaching unity at low temperature and decreasing(0.5) or less at high temperature [7].

The total conductivity was calculated from the equation:

$$\sigma_{total}(\omega) = (d/A)G \dots\dots\dots (3)$$

Where **d** is the thickness of the measured sample, **G** is the sample conductance, and **A** is the cross sectional area.

The dielectric constant ϵ was calculated from the equation:-

$$\epsilon = C/C_0 \dots\dots\dots (4)$$

where **C** the capacitance of the electrodes with dielectric, **C₀** is the geometrical capacitance of the sample without dielectric ($C_0 = \epsilon_0 A/d$, where ϵ_0 is the permittivity of free space and equal to 8.85×10^{-12} F).

The dielectric loss ϵ'' was calculated from the equation:-

$$\epsilon'' = G/\omega C_0 \dots\dots\dots (5)$$

The value of $(\tan\delta)$ can be calculated from the equation:-

$$\tan\delta = \epsilon'' / \epsilon' \dots\dots\dots(6)$$

Experimental Details

The material used as polymer matrix, epoxy resin type (EP10) and hardener type (HY-956) in ratio 3:1 for curing. One type of metal used in this work (aluminum powder) supplied by LDH chemicals Ltd (England) and practical size from (10 to 70 μ m). A weight amount of epoxy was mixed with aluminum powder is (20%, 30%, 40 and 50%) percentage.

For a.c measurements, samples were sandwiched between two gold electrodes and a programmable automatic LRC bridge (PM60304 Philips) were to measure the sample conductance G and the capacitance C directly. The measurements were carried out through at room temperature (298) K and frequency range (10^2 - 10^6) Hz.

Result and Dissection

A.c electrical properties of aluminum-epoxy composites were studied as a function of the constriction, and frequency, Fig.(2) show the relation between $\ln \sigma_{a.c}(\omega)$ and $\ln(\omega)$ for pure epoxy at room temperature (298) $^{\circ}$ C, and Fig.(3 a, b, c, and d)for epoxy/aluminum composites. It is clear from the figures that $\sigma_{a.c}(\omega)$ increased with the increase of frequency according to eq.(2) at room temperature. Increase of frequency increased a.c. conductivity by increasing the hopping of conducting electrons present in aluminum. At higher frequencies this hopping frequency could not match the applied field frequency.

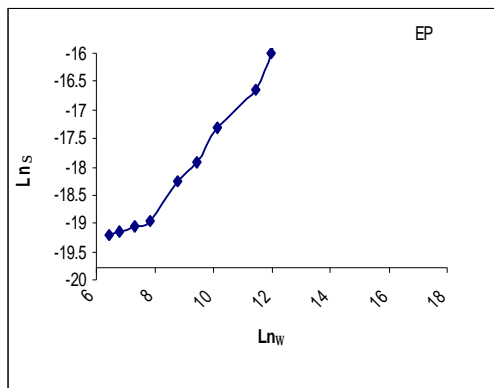
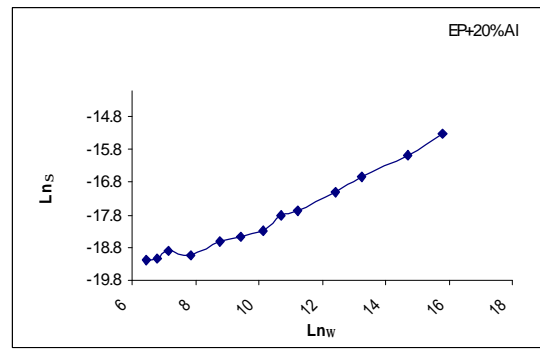
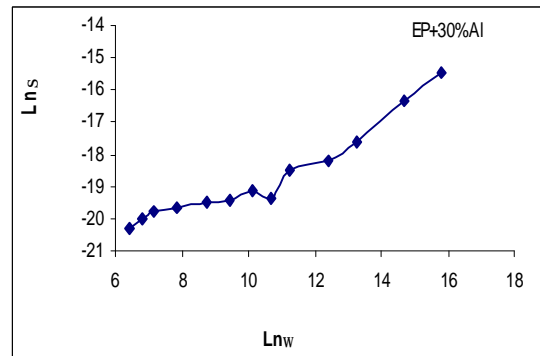


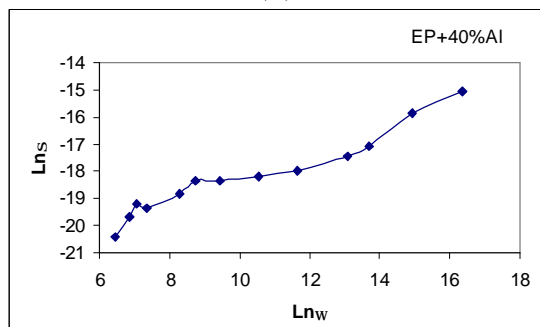
Fig. (2) Frequency dependence of the electrical conductivity for epoxy.



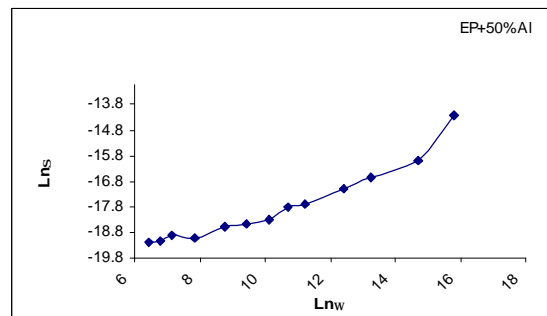
(a)



(b)



(c)



(d)

Fig.(3) Frequency dependence of the electrical conductivity for epoxy/aluminum composites of (a)20%,(b)30%,(c)40%,(d)50%.

Those figures have revealed that $\sigma_{a.c}(\omega)$ increased with increasing frequency. Hence, it is proposed that two factors influence $\sigma_t(\omega)$, which are ions motions and polymer backbone (main chain) motion. Furthermore ions motion is contributed at high frequency [9]. The increasing of $\sigma_t(\omega)$, at

low frequencies over (100-400) Hz is the attributed to the interfacial polarization, since the direct conductivity ($\sigma_{d.c}$) is significant at this region [10]. The rapidly increasing of $\sigma_t(\omega)$ with increasing frequency at the frequency greater than 10^3 Hz referred to the electronic polarization effect, and the conductivity is pure a.c conductivity $\sigma_{a.c}(\omega)$ in this region. At low frequencies where the applied electric field forces the charge carriers to drift over large distances. When frequency is raised, the mean displacement of the charge carriers is reduced and the real part of the conductivity, after reaching a certain critical frequency, f_c , follows the laws a.c. $(\omega) \sim (\omega)^s$ with $0 \leq s \leq 1$.

Characterizing hopping conduction. The critical frequency, f_c , has been found to be dependent on temperature and conductive filler volume fraction.

Hence the $\sigma_{a.c}(\omega)$ at high frequencies is proportional to the angular frequency (ω) according to the eq.(2). Values of the frequency exponent (s) were calculated for all the investigated composites from the slopes of the linear lines of $\ln \sigma_{a.c}(\omega) = f(\omega)$. It is observed that the frequency is found to have a pronounced effect on conductivity at lower temperatures. The obtained values of (s) ranged from (0.3 to 0.6) unit that indicate the correlated barrier hopping (CBH). According to the model, a.c conductivity $\sigma_{a.c}(\omega)$ can be explained in terms of the hoping of electrons between paries of localized states at the Fermi levels[11].Is the dominant conduction mechanism, and have a tendency to decrease with the increase of frequency and construction as indicated in Table (1).

Table (1)
Values of frequency exponent (S) and construction.

construction	s
EP pure	0.61
20% Al	0.48
30% Al	0.43
40% Al	0.41
50% Al	0.34

The variation of the dielectric constant ϵ with frequency was studied for Epoxy/Al composites at room temperature as shown in Fig. (4). The dielectric constant (is the ability of a material to store an electric charge), increased smoothly with an increase in the concentration of aluminum because it was attributed to interfacial polarization. An increase in the dielectric constant was observed with a decrease in the frequency because it can be attributed to the fact that the electrode blocking layers is dominated mechanism at low frequency region. Thus the dielectric behavior is affected by electrodes polarization. At high frequency the dielectric signal is not affected by electrodes polarization. When the frequency is increased, the dipole cannot rotate sufficiently rapidly, so that their oscillations lag behind those of the field. As the frequency is further raised the dipole will be completely unable to follow the field and the orientation polarization ceases, so decrease approaching a constant value at high frequencies due to the interfacial polarization only [12].

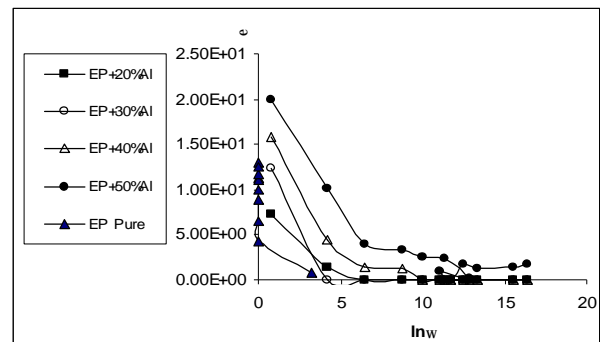


Fig.(4) Frequency dependence of the dielectric constant(s) fo EP Pure and EP/Al composites of different ratio.

The dissipation factor $\tan\delta$ (is the degree of dielectric loss) .In general, dissipation factor values for composites with higher concentrations of aluminum were greater than that of lower volume content of aluminum was attributed to interfacial polarization. Also, the dissipation factor showed in Fig. (5) increased with a decreased in frequency at room temperature. In low signal frequency range, when the electrical dipoles are an able to follow the variation of the electric field, the dissipation factor decreases with the increases of signal frequency [13].

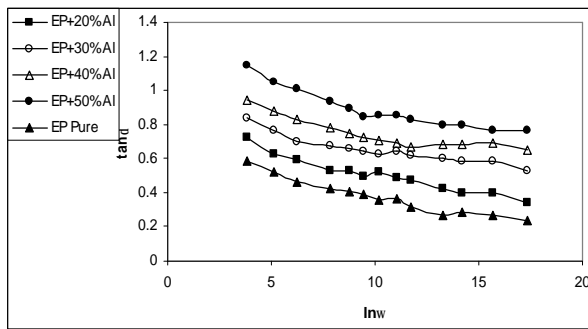


Fig. (5) Frequency dependence of the dissipation factor of $\tan \delta$ for EP Pure and EP/Al composites of different ratio.

Conclusion

- 1-The a.c conductivity increases as the increase of frequency, and the frequency exponent(s) decreases as the increase of frequency.
- 2-The dielectric constant decreases with the increases in frequency, and dissipation factor increases with a decrease in frequency, and dissipation factor increase with increase of concentration of aluminum.
- 3-The correlated barrier hopping (CBH) is the a.c conduction mechanism in the composites samples

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الخلاصة

يتناول هذا البحث دراسة خصائص العزل للإيبوكسي النقي والمترابك المتكون من الإيبوكسي / الألمنيوم. تم قياس التوصيلية الكهربائية $[\sigma(w)a.c]$ للمترابكات بنسبة 20 %، 30 %، 40 %، و 50 % وبمدى تردد بين $(10^2 - 10^6)$ هرتز في درجة حرارة الغرفة ووجد بان التوصيلية تزداد بزيادة التركيز.

لقد وجد لكل العينات ثابت العزل الحقيقي (ϵ) وعامل النقص ($\tan \delta$) حيث ان ثابت العزل الحقيقي (ϵ) يزداد بشكل

قليل مع زيادة تركيز الالمنيوم المضاف والسبب يعود الى بينية الاستقطاب ويقل مع زيادة التردد. بصورة عامة فان قيم عامل الفقد ($\tan\delta$) للمركبات المحتوية على نسبة عالية من الالمنيوم تكون اعلى من تلك التي تحتوي على نسبة قليلة من الالمنيوم وقد لوحظ بان عامل الفقد يزداد مع تناقص التردد. وجد بان التوصيلية المتناوبة تزداد مع زيادة التردد وفق العلاقة $S(w)=Aw^s$ ويقوم للعامل الاسي (s) اقل من الواحد لكل العينات وتتناقص مع زيادة التردد وتقع بين (0.3-0.6) وحدة، وجد بان الية التوصيل السائدة هي نموذج القفز فوق حاجز التنطط (CBH).