Effect of Water Absorption on Some Electrical and Dielectrical Properties of Epoxy Resin Reinforced with Chopped Carbon Fibers

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Abstract

In this research, we study a.c electrical conductivity ($\sigma_{a.c}$ (ω)) and dielectric properties (dielectric constant (έ), and dissipation factor (tanδ)), with frequency change (10^2 -10^7 Hz) for the epoxy resin and composites reinforced with chopped carbon fiber ,prepared by hand lay –up method (EP +5% CF), (EP +10% CF), (EP +15% CF), (EP +20% CF), Were measured electrical conductivity of epoxy and composites with frequency change and at room temperature the results showed a.c electrical conductivity dependence with frequency relationship σ (ω)=Aω^s to the values of the exponential factor (s)of less than one all samples, and adopted the theory of filtration for the interpretation of data conductivity for composites reinforced with carbon fibers was selected epoxy+20% C.F showed a higher electrical conductivity (13.6 * 10^{-7} $(ohm.cm)^{-1}$).

The composites were immersed in water at room temperature for 6, 10,15,20,27 days. The results showed that the electrical conductivity of (σ (ω)) increasing with increasing weight gain(M%) due to water absorption. Itwas found that dielectric constant increases when the percentage moisture absorption increase, whereas dielectric constant (έ) is dependent on capacity of composite materials.

Keywords: epoxy, carbon fibers, electrical conductivity, dielectric properties.

Introduction

Composite material may be defind as any substance which is made by physical combining of two or more existing materials to produce amultiphase system with different physical properties from the starting materials but in which the constituents retain their identity [1].

A.C Conductivity is one of the studies done on solids in order to characterize the bulk resistance of the material sample .Measurment of a.c conductivity can be done by different techniques. The currently used technique is the complex impedance spectroscopy. This study also gives information on electrical properties of materials and their interface with electronicallyconducting electrodes [2].

The epoxy resins have been used extensively as adhesives and matrix resins for fibrous and particulate reinforced composite materials, because they have favorable properties such as high modulus, low creep and reasonable performance at elevated temperatures [3]. Epoxy resins have been used as an encapsulation of semiconductor devices and chips, as the capacitance of semiconductor chips or devices increases the heat dissipation caused increases the cracks produce on epoxy surface[4]. Epoxies are widely used in insulation, such as electrical machinery, switchgear and bushings in transformers (Wuand Tung 1995). Polymers have a very low concentration of free charge carriers, and thus are non conductive and transparent to electromagnetic radiation. Fibrous polymer matrix composites(FPMC) have become that most important categories of the composite materials, because these composites have excellent flexibility to modify there physical, thermal and electrical properties[5] The FPMC Materials can be classified according to the type of fibrous materials into two types continuous fiber reinforced composites and discontinuous fiber reinforce composites [6].

Carbon Fiber Reinforced Polymers (CFRP) are characterized by many properties: light weight, high strength, very high modulus elasticity, high fatigue strength, good corrosion resistance, very low coefficient of thermal expansion, and high electric conductivity [7].

Water absorption can be defined as the amount of weight gain experienced in a polymer after immersion in water for a specified length of time under a controlled environment. Water absorption for long time and at high temperature can be problem with polymeric resin and natural fibers because it can lead to swelling [8].

In 2002 Allaouiet al [9]Studied the effect of carbon walled nano-tubes on the dielectric properties of epoxy matrix composite.The result exists increasing in conductivity by nine order of magnitude at 4% weight fraction of carbon Nanotube.

In 2005 the A.C.electrical properties of fiber glass\epoxy composites, studied by Al-shabander [10] was found that the A.C. electrical conductivity increase with increasing frequency, glass fibers wt%, water absorption, and temperature.

In 2006 the effect of water on electrical properties of polymer composite with cellulose fibers were studied by Notingher and friends[11] they were found that the immersion in water of polymer composite samples affects the electrical behaviour of composites as samples of polymer composites with treated cellulose fibers keep better resistivity values after immersion in water.

Dielectric and Electrical Measurement

Dielectric constants were calculated at room temperature.The Real Part of dielectric Constant (Permitivity), $\acute{\varepsilon}$, was calculated from the following equation.

$\acute{\varepsilon}= C_p / C_0$ ....(1)

Where C_p and C_0 are the Capacity of the electrodes with and without dielectric respectively ; C_0 is given by:

$C_0 = [0.08854\ A/d]$ ....(2)

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

The dielectric loss (imaginary Part of dielectric Constant), ε'' has been calculated by following equation [12]using the measured value of dissipation factor ($\tan\delta$)from RLC Bridge.

$\varepsilon^{//} = \acute{\varepsilon} \tan \delta$ ....(3)

the A.C conductivity; $\sigma_{A.C}$, could be calculated by the following equation [13]

$\sigma_{a.c} = \omega\, \varepsilon_0\, \varepsilon^{//} = \omega\, \varepsilon_0\, \acute{\varepsilon} \tan \delta$.............................(4)

Where, ω, is the angular frequency and equal to ($2\Pi f$) where,f,is the frequency ,

Afrequency dependence on a.c conductivity ($\sigma_{a.c}(\omega)$)has been observed in many amorphous semiconductors and insulators both inorganic and polymeric material, has the form [14]

$\sigma_{a.c}(\omega)= A\, \omega^s$ ….. (5)

where s is the exponent($0 < S < 1$)and A is the proportional factor.

The Total measured conductivity ($\sigma_t(\omega)$) at a given frequency (ω) is separable into d.c and a.c components, namely.

$\sigma_t(\omega)=\sigma_{a.c}(\omega)+\sigma_{d.c}$ .... (6)

$\sigma_t(\omega)=A\omega^s +\sigma_{d.c}$ .... (7)

where $\sigma_{d.c}$ is the d.c conductivity, $\sigma_{a.c}(\omega)$ is the high frequency dispersion of $\sigma_{a.c}$, which appearsat low frequencies is due to d.c .conductivity and that $\sigma_{a.c}$ takes off from $\sigma_{d.c}$ at an angular frequency $\omega=1/\tau$ where τ is the relaxation time for A.C Conductivity Fig. (1) illustrates the variation in conductivity, There are two Dispersions, the first is due to D.Cconductivity (the Plateau b) and the low frequency dispersion (a) is attributed to the electrodes polarization effect while the high frequency dispersion (c) is corresponds $\sigma_{a.c}(\omega)$ .Thus in this region $\sigma_{a.c}(\omega)$ is proportional to ω^s, and usually the exponents(s) is estimated from the slope of high frequency dispersion [15].

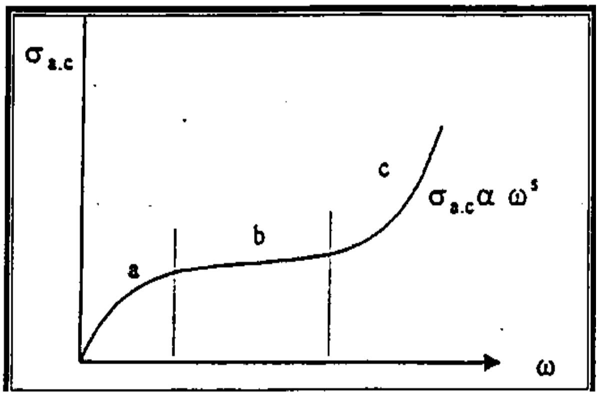


Fig. (1) The variation of A.C. conductivity with angular frequency.

Experimental Work

1- Materials;-

1-1 Epoxy resin

Epoxy resin was used as a matrix (Mastertop 1230 plus), adhesive grade room temperature supplied by Iranian BASF construction chemical .the ratio of hardener to epoxy used in this study are approximately [1:2].

1-2 Carbon fibers

Carbon fibers were used in this study to reinforce the Epoxy which was supplied by Grazer company Germany. The length of carbon fibers chopped (short) 1-2 cm, fiber diameter 7-8μm, and with density 1.75 gm/cm^3.

2- Sample Preparation:-

Hand lay-up technique was used to prepare sheets of composites. Weight amount of Epoxy was mixed with carbon fiber in different percentages(5%,10%.15%,20%) and the samples were cut as disc shaped panels of 3 cm diameter and have a fixed thickness of 3mm, for electrical and dielectrical measurements. The test samples of neat epoxy resin and epoxy/chopped carbon fibers composites were immersed in water for 6,10,15,20,27 days. The samples removed from water every 24 hours and weight by using an the balance (metter H35 AR)of accuracy 10^{-4}. The weight gain percentage (M%) was calculated by using the following equation

$$M\%=[(W_{(t)} - W_0) / W_0 \times 100] \quad ....(8)$$

Here, W(t)is the total weight at time (t) and W_0 is the reference dry weight of the composite .

3- The A.C characteristics measurements:-

Studying the A.C electrical characteristics may provide valuable information about the conduction mechanisms of epoxy-carbon fiber composites. The values of these effects can be understood from the variation of capacitance, tanδ and electrical conductivity with frequency.The range of the frequency that of concern to this work is 10^2-10^7 Hz at room temperature.

To study and measure the effect of fiber wt%, and frequency of applied electric field on theA.C electrical conductivity, έ, and tanδ of fiber epoxy composite and the guard electrode method has been used. As shown in Fig.(2) the specimen holder is situated in temperature cotrolled oven (Hereaus Electronic). The high and low specimen holder terminal was connected to multi-frequncy LCR Meter(models HP-4274 A and HP-4275 A)and the third specimen holder terminals is connected to earth .The HP-4274A and HP-4275A are enable to measure several parameters, which are them, the specimen capacitance, dissipation factor, resistance, and the phase angle with an accuracy of (0.1 %) and resolution of (5.5). The HP-4274A covers a frequency range of (100Hz-100KHz) in 11 spots while the frequency covered by HP-4275A has10 spots ranging from (10KHz-10 MHz).

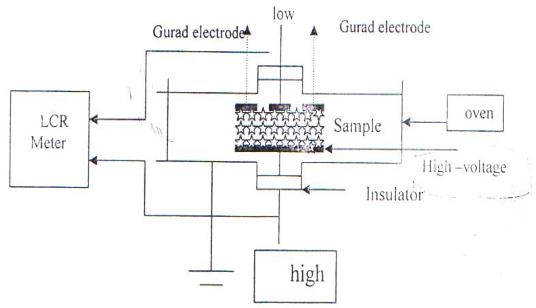


Fig. (2) Schematic demonstrating the type of electrical connection.

Results and Discussion

1-Frequency dependence of electrical conductivity ($\sigma_{a.c}$) ,dielectric constant (έ),and dissipation factor(tan(δ)):-

A f requency dependence on a.c conductivity ($\sigma_{a.c}(\omega)$) has been observed in many amorphous semiconductors and insulators both inorganic and polymeric material, has the form

$\sigma_{a.c}(\omega) = A\,\omega^s$, where, s is the exponent, A is the proportional factor.

1–1 The effect of chopped carbon fibers ratios on $\sigma_{a.c}$(w),έ,and tan(δ) of epoxy resin:-

Fig.(3) shows the variation of the A.C conductivity ($\sigma_{a.c}$) with frequency for epoxy-carbon fiber composites for different wt% fibers at room temperature (303 K), while it increased rapidly above 10 KHz to reach the value of 12.4×10^{-8} $(ohm.cm)^{-1}$ at for epoxy. In this frequency region the $\sigma_{a.c}(\omega)$ is due to two main processes those are called ions motion and the transport near or at the interstitial surface between carbon fibers and epoxy resin. The increasing of $\sigma_{A.C}(\omega)$ in high frequency region can be related to the electronic polarization The electrical conductivity increase with increasing the weight of carbon fibers, These electrical conductivity of EP+20% chopped carbon fibers, show similar values with semiconductor materials, The values of electrical conductivity for epoxy and composites at 100Hz given in Table (1).

Table (1)
The values of $\sigma_{a.c}$ (ω)for Epoxy and composites

Material	*Ln $\sigma_{a.c}$ (ohm.cm)$^{-1}$*
EP	-21
EP+5% C.F	-20.3
EP+10%C.F	-19.1
EP+15%C.F	-17.5
EP+20%C.F	-16.5

The dependence of the dielectric constant on frequency for epoxy resin show adebye-type relaxation. Dielectric constant values decrease with increasing frequency. The variation of έ as a function of frequency at room temperatures (303K) for EP reinforced by carbon fibers. The result shows that the values of έ are affected by the adding of reinforcement material. such έ is shown highly frequency depended at the frequency range over 10^2-10^7 Hz for the carbon fiber. Figs. (4), This result can be explained by pohl and Pollack [16] proposal, they are proposing that an intra-chain transport mechanism involving polarnos. This mechanism does allow an explanation of usually high dielectric constants and associated phenomenon of nomadic polarization observed in many polymer composites. Also the electrode blocking layers mechanism may be contributed and the electrode polarization is exists [17]. Figs.(5) show the variation of dissipation factor (tan(δ)) as a function of frequency at room temperature (303K) for EP reinforced by chopped carbon fiber. The result shows that the strong dispersion occurs in EP/C.F, This result has been attributed to strong carrier polarization arising from high densities of low mobility charge carriers [18]. The values of tan (δ) are increased with increasing carbon weight fraction.

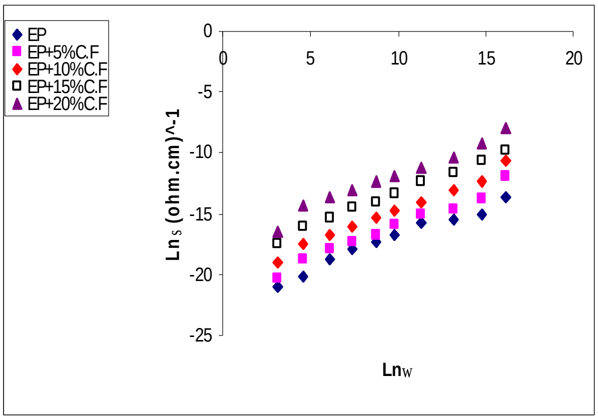


Fig. (3) The variation electrical conductivity with frequency for epoxy and epoxy -carbon fiber composites at (303K).

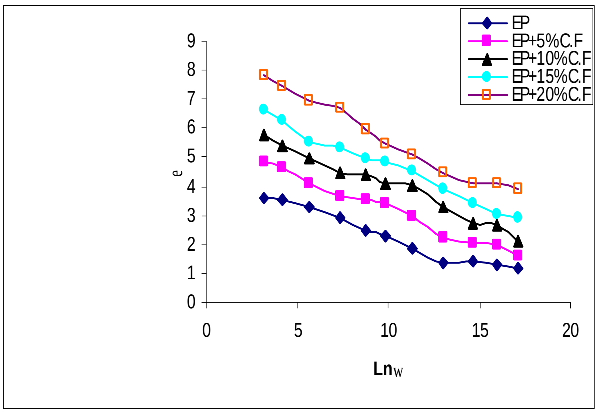


Fig.(4) The variation of έwith ln the (w) for epoxy and epoxy–carbon fibers composites at 303 K.

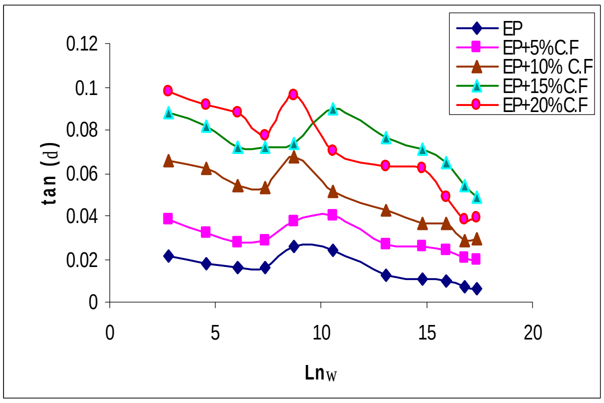


Fig. (5) The variation of tan (δ) with ln (w) for the epoxy and epoxy-Carbon fibers composites at 303 K.

1-2 The Effect of water Absorption on $\sigma_{a.c}$ (w), έ, and tan (δ) of EP/chopped carbon Fibers:-

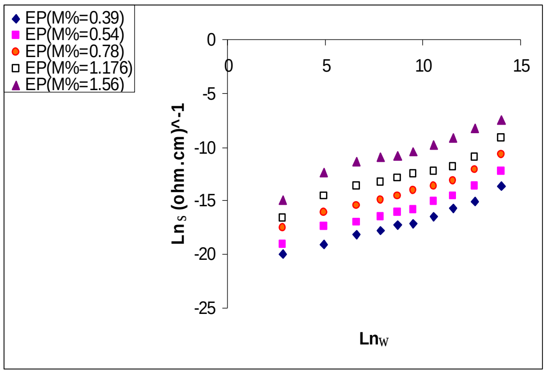
Weight gain (M %) have been calculated from equation (8), and the values are listed in Table (1). The structure factors which favor diffusion also tend to increase absorption (or swelling). For this reason crystalline or cross linked polymers are specified where volume stability is necessary [19]. Diffusion in polymeric molecules differs from that of atoms or ions in two ways, first the macromolecules are large and cumbersome because they coil and link, their entanglement inhibits significant movement, past other molecules. Second the use of polymeric solids is confine relatively low temperatures, also the type and nature of polymers used are very important in the diffusion process.

Water has a low electrical conductivity, but this increases significantly upon salvation of a small amount of ionic material such as hydrogen chloride or any saltThus the risks of electrocution much greater in water with the usual impurities not found in pure water. (It is worth noting, however, that the risks of electrocution decrease when the impurities increase to the point where the water itself is a better conductor than the human body. For example, the risks of electrocution in water are lower than in fresh water, as the sea has a much higher level of impurities, particularly common salt, and the main current path will seek the better conductor [20]. An important feature of water is its polar nature. The water molecule forms an angle, with hydrogen atoms at the tips and oxygen at the vertex. Since oxygen has a higher electronegativity than hydrogen, the side of the molecule with the oxygen atom has a partial negative charge. An object with such a charge difference is called a dipole. The charge differences cause water molecules to be attracted to each other (the relatively positive areas being attracted to the relatively negative areas) and to other polar molecules. [21], the increase of percentage water absorbation executes increase the electrical conductivity for epoxy pure and composites.

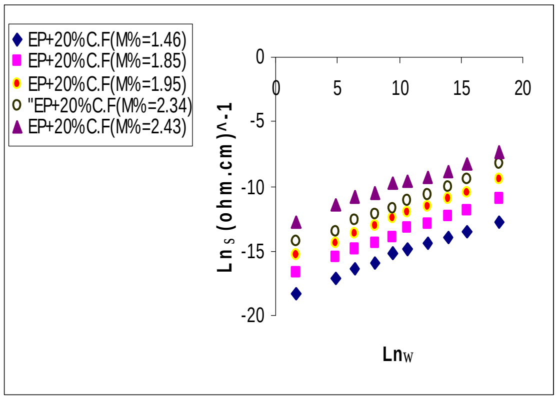
The valus of exponent (s) were estimated from the slope of the curves of Ln $\sigma_{A.C}$ Versus Ln (ω) , are shown in Table (1). Table (1) reveals that the values of s were increased with increasing obsorpation of water Fig.(8).

Table (2)
The values of weight gain(M%)for epoxy and epoxy with 20%chopped carbon fibers and The values of exponent (s) for EP and EP+ 20%C.F.

M% (EP+ 20%CF)	*M% (EP)*	*S(EP+20% C.F) M% (EP)*	*S (EP)*	*T (days)*
1.46	0.39	0.57	0.5	6
1.85	0.54	0.59	0.55	10
1.95	0.78	0.61	0.56	15
2.34	1.17	0.64	0.58	20
2.43	1.56	0.67	0.59	27

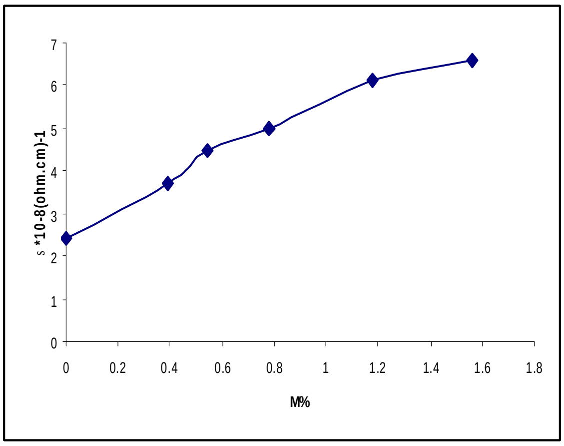


(a)



(b)

Fig (6) (a) The variation electrical Conductivity with frequency for Epoxy pure at (303K)
(b) The variation electrical Conductivity with frequency for Epoxy +20%carbon fibers Composites at (303k).



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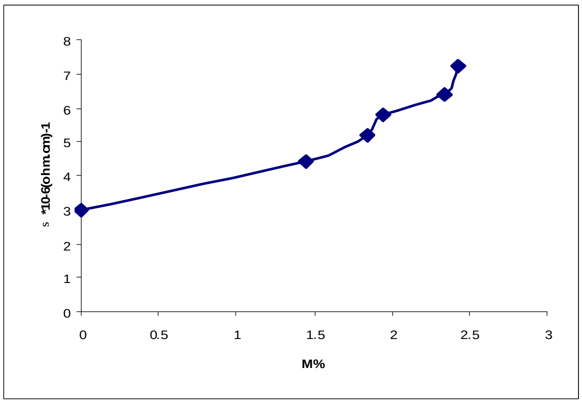
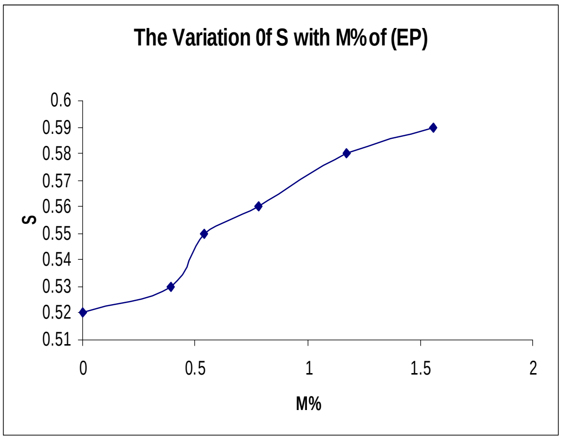
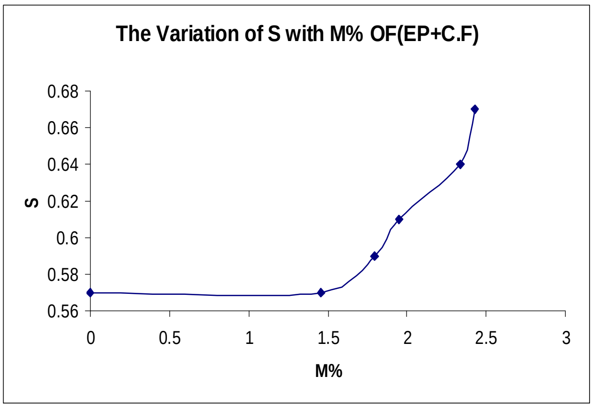


(b)

Fig. (7) (a) The variation electrical Conductivity with water Absorbency percentage at Frequency100KHz of epoxy.
(b) The variation electrical Conductivity with percentage of The water absorbency at Frequency100KHz of epoxy +20% Chopped carbon fibers.



(a)

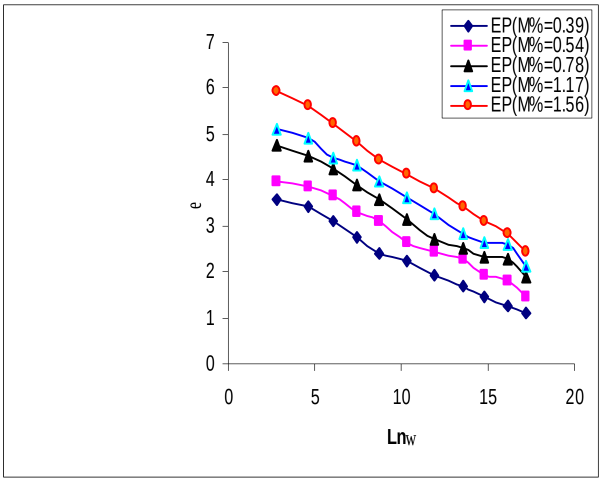


(b)

Fig.(8) (a) The variation of (s) with epoxy at (303K)
(b) The variation of(s) with epoxy-20%carbon fiber composites at (303K).

The Dielectric constant values inecrease with increasing percentage change of the water absorption of the epoxy resin and epoxy composites, Fig.(9) show the varation of έ as afunction of frequency at room temperature for EPComposites are reinforced by chopped carbon fibers, the result shows that the values of έ are affected by the adding of reinforcement material. The relative static permittivity of a solvent is a relative measure of its polarity. For example, water (very polar) has a dielectric constant of 80.10 at 20 °C while n-hexane (very non-polar) has a dielectric constant of 1.89 at 20 °C [22] .

Fig. (10) show the variation of dissipation factor (tan (δ)) as afunction of frequency at room temperature (303k) for EP and EP+20% C.F. The dissipation factor increase with increasing percentage change of the water absorption of epoxy resin and epoxy reinforced with chopped carbon fibers. The loss tangent peaks appearing at a characteristic frequency suggest the presence of relaxing dipoles in all the samples, the peak shifts towards higher frequency side suggesting the speed up the relaxation time.



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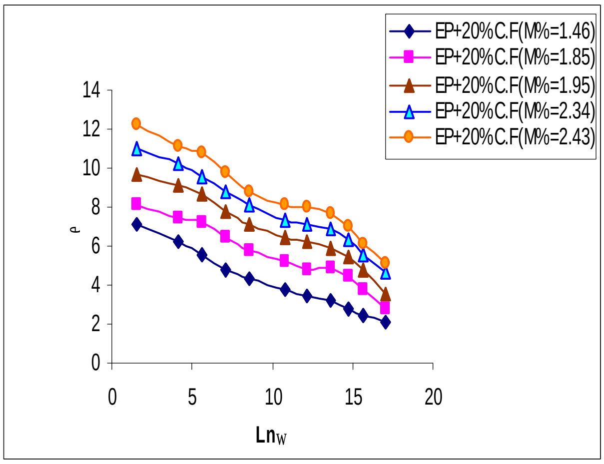
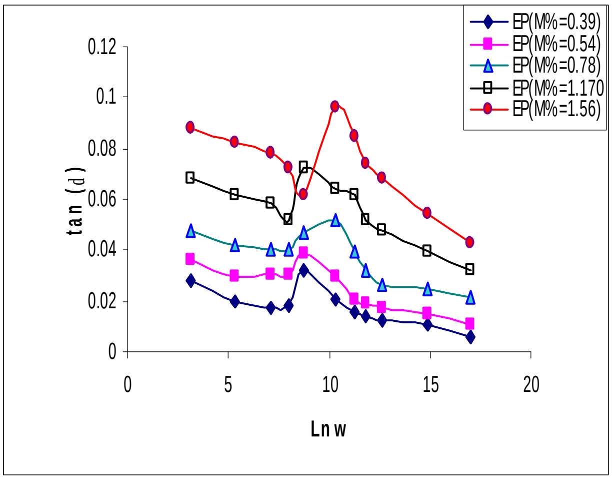


(b)

Fig (9) (a) The variation of έ with ln (w) for the epoxy
(b) The variation of έ with ln (w)for the epoxy-20%carbon fibers Composites.



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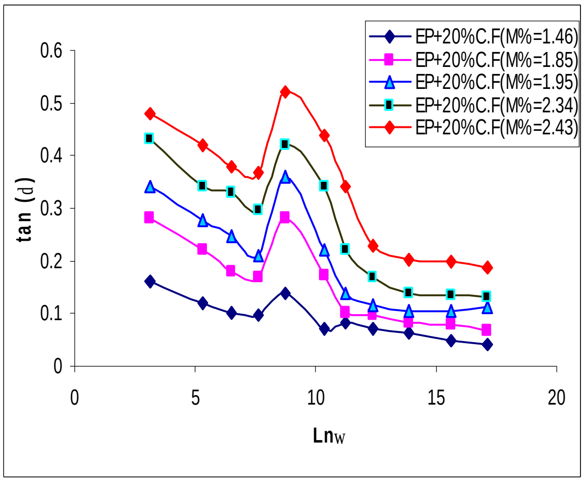

(b)

Fig. (10) (a) The variation of tan(δ)with Ln (w) for the epoxy resin
(b) The vatiation of tan (δ) with ln (w)for epoxy+20% carbon fibers composites

References

[1] Navin Chand and Deepak Jain "Evaluation of a.c conductivity behavior of graphite filled polysulphide modified epoxy composites", Vol.27. No.3, June, pp. 227-233, (2004).

[2] R. Padmd Suvrna, K. Ragha "Asimple technique for a.c conductivity measurement", Vol.25, No.7, pp.647-651, (2000).

[3] G. W. Yeager, J. M. Anostario, G. D. Merfled, M. T. Takemori, M. L. Todt, G. A. Hutehins, and Info. series", G.E. Research and Development center. (USA), (2000).

[4] Mr. Jeremy Hierhoizer, Assistant Professor of Aviation Technology, Purdue University, (2007).

[5] M.O.W.Richardson, "Polymer Engineering Composites", Applied Science LTD, London(1984).

[6] B.D.Agarwal, and L.J.Broutman" "Analysis and performance of fiber compsites", John Wiley and Sons. Inc. NewYork (1980).

[7] Z.Z hano and J.Gou "Improved relardancy of thermoset composites modified with carbon fibers" sci, technoi. Adv. mater 10 (2009).

[8] ComynJ."Polymer Permeability" (Elisiver Appl.Sci. Publishers London, (1985).

[9] A.Amaoui,S.Bai,H.M.Cheng and J.B.Bai, "Composite Science and Technology", Vol.62, pp.5691-5692, (2002)

[10] Ban Mazain Al-shabander "Study the electrical properties for Epoxy resin reinforced with fiber glass", department of Physics, college of scscience, university of Baghdad.(2005)

[11] P.V.Notinghes, D.Panaltescu, Z.Vuluga, M. Iorga, H. Paven, "Journal of Optoelectronics and advanced materials", Vol.8. No.2, pp.687-689, (2006)

[12] Basil A.Abdulla,Kays Majadi & Araxsy , A.Tosonian, Iraqi, J.Polymers Vol.5, No.1, pp.1-14, (2001).

[13] H.Christopher,"PolymerMaterials" 2ed , Mac Milan Education.Ltd., (1989).

[14] A.K.Jonscher. "Thin Solid Films", Vol.36, (1976).

[15] P.Extance, S.R.Elliot and E.A.Davis, "Phys.Rev.B", Vol.32, No.12 (1985).

[16] H, A.Pohl and M.Pollok, "J.Chem.Phys.", Vol.63, pp.2980 -2981, (1975).

[17] T.Jeevanada, and Siddaramaiah "Euro. polym.J.", Vol.39, pp.569-570, (2003).

[18] A.K.Jonscher, "Thin solid films" Vol.11, pp.206-207,(1980).

[19] Crark J. and Park G.S.,"Diffusion in polymers", Acadimic press Inclondon, 1968.

[20] Phil sehewe, James Riordon, and Ban Stein "Awater molecules chemical formula is Really Not H_2O", Physics News apdate (31 Jul, 2003).

[21] R.Sathamothy, S.Narayandass, C.Balas, and D.Mangala, Phys.Stat.Sol.Vol.117, pp.495-496, (1990).

[22] D.R. Lide, Ed. *CRC Handbook of Chemistry and Physics, 85th Ed.* (2004). CRC Press. Boca Raton.pp.8-14.

الخلاصة

تم في هذا البحث دراسة التوصيلية الكهربائية المتناوبة ($\sigma_{a.c}(\omega)$) وخواص العزل الكهربائي (tanδ,έ) مع تغير التردد (10^2 -10^7 Hz) لراتنج الايبوكسي ومتراكباته المدعمة بالياف الكاربون المقطعة والمحضرة بطريقة التشكيل اليدوية (EP+5%C.F)، (EP+10%C.F)، (EP+15%C.F)، (EP+20%C.F) تم قياس التوصيلية المتناوبة للايبوكسي ومتراكباته في مدى التردد وفي درجة حرارة المختبر اظهرت النتائج اعتماد التوصيلية على التردد وفق العلاقة $\sigma(\omega)=A\omega^s$ بقيم للعامل الاسي اقل من الواحدلكل العينات, واعتمدت نظرية الترشيح لتفسير بيانات التوصيلية لمتراكبات الايبوكسي المدعم بالالياف الكاربون. كما تم دراسة تأثير التردد في قيمة ثابت العزل الكهربائي وفقد العزل فقد وجد بان السماحية (έ) تقل في حالة التقوية بالياف الكاربون المقطعة، تم اختيار المتراكب ايبوكسي –20% الياف الكاربون المقطعة الافضل من بين المتراكبات باعتباره اظهر اعلى توصيلية كهربائية ($13.6*10^{-7}$ $(ohm.cm)^{-1}$). تم تغطيس المتراكبات في الماء بدرجة حرارة الغرفة لمدة 20,1510,6 , 7, 2 يوم، اظهرت النتائج العملية ان التوصيلية الكهربائية (σ(ω)) تزداد بزيادة التردد وزيادة النسبة المئوية لامتصاصية الماء بينما ثابت العزل (έ) يزداد عند زيادة النسبة المئوية لامتصاصية الماء اعتمادا على السعة الكهربائية للمواد المتراكبة.