Effect of Nanocomposites TiO_2 addition on the Dielectric Properties of Epoxy resin

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Abstract

Sheets of Epoxy (EP) resin with addition of TiO_2 of grain size (1.5μm, and 50nm) and weight percentage (1%, 3%, and 5%) were prepared. Discs of 20mm diameter and 3mm thickness were cut for dielectric measurements. Dielectric properties (dielectric constant, dispassion factor and electrical conductivity) over the frequency range 10^2 -10^6 Hz were measured.

Comparison was made between the effect of micro and nano particles of TiO_2 on the dielectric properties of EP composites with different weight percentage. Epoxy composites with micro sized particles of TiO_2 were observed to have the better values of dielectric properties.

Keywords

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تأثير متراكبات النانو بإضافة TiO_2 على خواص العزل لراتنج الأيبوكسي

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الخلاصة:

في هذا البحث تم تحضير صفائح راتنج الايبوكسي Ep باضافة ثنائي اوكسيد التيتانيوم TiO_2 بمقاس حبيبي (1.5μm, 50nm) وبنسب وزنية مئوية مختلفة (1%, 3%, 5%). تم قطع اقراص بقطر 20mm وسمك 3mm من المادة المحضرة ولذلك لغرض القياسات العزلية. الخواص العزلية (ثابت العزل وعامل الفقد والتوصيلية الكهربائية) اجرت القياسات لمدى من الترددات بين 10^2 -10^6 Hz.

تم كذلك مقارنة تأثير الجسيمات TiO_2 من ابعاد النانومتر والمايكرمتر على الخصائص العزلية من متراكبات الايبوكسي وبنسب وزنية مختلفة. وتبين من النتائج ان الجسيمات TiO_2 بابعاد المايكرو متر ذات خصائص عزلية افضل

Introduction

Epoxy resin is an indispensable material for insulation systems in heavy apparatuses such as switchgear .It is reinforced by micro-scale fillers or fibers to obtain essential properties for heavy apparatuses. Recent approaches to meeting an ever-increasing demand for high performance material have tended to concentrate on polymer nano-composites, and there have been a large number of related reports. However, the paucity of studied on the polymer/nano-scale filler interface has encouraged us to investigated it because fundamental study is essential for the polymer nano-composites technology to be applied to insulating materials .The resin was selected because it was benign (i.e. without other fillers or diluents), had a low initial viscosity, and a glass transition below 100 ºC[1,2].

Dielectric properties of epoxy-resin specimens can be improved (e.g. resistance to surface degradation) by the use of nano-sizedalumina fillers. The improvement was seen to be even more marked if the nanometric fillers were pre-processed before use **[**3]**.**

Polymer nanocomposites are already a part of many important of worldwide businesses: automotive (molded part in cars), electronics and electrical engineering, household products, packaging industry, aircraft interiors, appliance components, security equipments. Among many nanocomposite precursors.TiO_2 nanopowder is increasing labeling investigated due to its special properties .Among many nanocomposite precursors, TiO_2 nanopowder is increasingly being investigated because it is non-toxic, chemically inert, low cost, has a high refractive index[3].

Polymer matrix ceramic filler composites such as EP-TiO_2, EP-Al_2O_3 and EP-SiO_2 composites are receiving increased attention due to their interesting electrical and dialectical properties .Integrated capacitors, acoustic emission sensor sand and electronic packing are some potential properties [3,4]. TiO_2 has been the subject of many studies due to its remarkable optical electronic properties [5]. Epoxy resin was chosen as a matrix due to its good adhesion with the fillers, having enhanced thermal stability resistance to chemical attack and resistance to the degradation [6, 7]. New fields of dielectronic with high dielectronic constants appeared in recent years with the preparation of ceramic nanoparticles such as nano TiO_2, Al_2O_3, MgO, and ZnO for polymer composites [8, 9, and 10].

This work deals with studying the dielectric behavior of two different types of epoxy composites with nano TiO_2, and micro TiO_2 over the frequency range 10^2-10^6 Hz.

Electrical and dielectric measurements

The empirical relation for the frequency dependence of a.c conductivity is given by[11,12] :

$$\sigma_{a.c}(\omega) = A_1\omega^s \qquad \text{………… (1)}$$

Where:

A_1 is constant, **ω** is the angular frequency ,and**(s)** is a function of temperature and is determined from the slope of a plot Ln $\sigma_{a.c}(\omega)$ versus Ln **ω**, then **(s)** is not constant but decreases with increasing temperature, usually 0<s <1 it approach as unity at low temperatures and decrease to (0.5) or less at high temperatures [13].

The total conductivity was calculated from the equation:

$$\sigma_{total}(\omega) = (d/A)G \qquad \text{……………….(2)}$$

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

The dielectric constant **ε** was calculated from the equation:-

$$\varepsilon = C/C_\circ \qquad \text{………. (3)}$$

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

The dielectric loss **έ** was calculated from the equation**:-**

$$\acute{\varepsilon} = G/\omega C_\circ \qquad \text{………………….. (4)}$$

Then dissipation factor (tanδ) the degree of dielectric loss can be calculated from the equation:-

$$\tan\delta = \acute{\varepsilon}/\varepsilon \qquad \text{……………… (5)}$$

Sample preparation method

Epoxy (EP10) and hardener type (HY-956) were used in this study in ratio of 3:1 for curing with nanoTiO_2 (50nm) and micro TiO_2 (1.5µm) to form nano and micro composites with different TiO2 weight percentage (1%, 3%, and 5%).Careful mixing of EP with TiO_2 was needed to minimize voids and clustering of the particles.The specimane were cut dimensions (250mm*250mm*3mm).

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

Results and Discussion

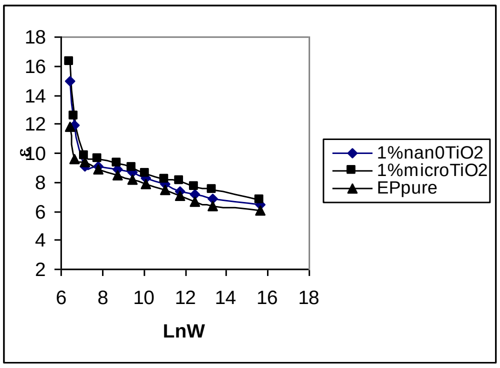
Dielectric constant values are calculated using eq.(4)for Epoxy(EP) pure and EP composites with micro and nanoTiO_2 particles as shown in Table (1).

Table (1): Dielectric constant values for EP and EP/TiO_2 composites (nano and micro)

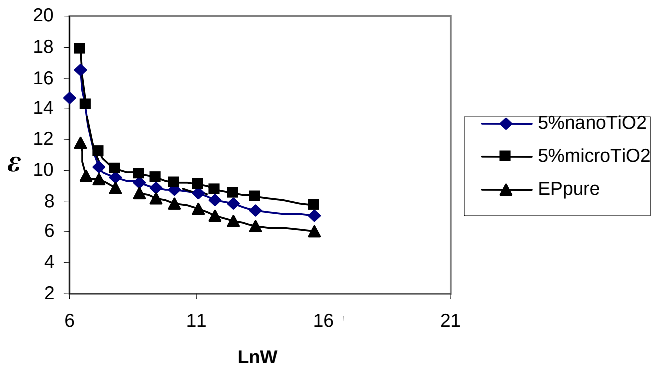
Samples	ε at 10^3 KHz
EP PURE	12.2
EP+1% micro T iO_2	16.5
EP+3% micro T iO_2	17.6
EP+5% micro T iO_2	18.3
EP+1% nano T iO_2	15.1
EP+3% nano T iO_2	15.5
EP+5% nano T iO_2	16.3

Figure (1) shows relative permittivity plots obtained for the three micro composites (EP resin and 1%TiO_2 micro, and nano composites) at room temperature (298ºK) and high frequencies. The micro filled composites has the higher real relative permittivity (ε) due to the high relative permittivity of the filler .It is instructive to compare the real part of the relative permittivity of three composites at 1KHz where the low frequency effects do not dominate. At room temperature, EP resin exhibits a real relative permittivity of 12.2.compared to 13.3 for nano composites; the nano particles have a profound effect on the dielectric behavior of the nanocomposites. They appear to restrict end-chain or side-chain movement of the epoxy molecules.

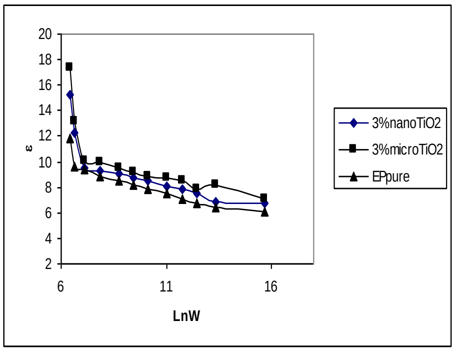
Composites of polymer matrixes fine ceramic particles are considered as disordered systems with electrical and dielectric performance related directly to the type, weight, size, shape of the fillers [5, 6, 9, 10] . Other workers reported higher real relative permittivity (ε) for EP/micro 1%TiO_2 .However; EP nanocomposites exhibit lower measured values of (ε) at high frequency which is less than that of the pure resin matrix (EP)[5].



(a)



(b)



(c)

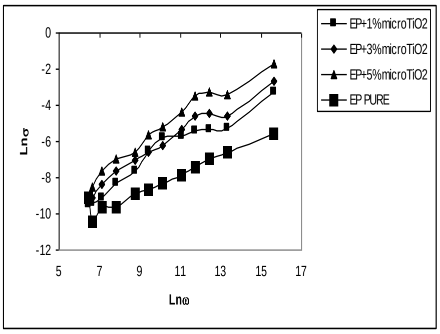
Fig (1 a,b,c)::Frequency dependence of ε for EP Pure and EP/TiO_2 composites (nano and micro)

Fig (2) shows the relation between ln $\sigma_{a.c}(\omega)$ and ln(ω) for pure epoxy and epoxy TiO2(nano and micro composites) at room temperature (298) ̊K .At very low frequencies, the dielectric behavior the base resin and micro composite become very similar as electrode effects dominate. It is clear from the figures that $\sigma_{a.c}(\omega)$ increases with the increase of frequency(according to eq.(2)) at room temperature. The highest conductivity for EP Pure and EP/TiO_2 composites (nano and micro) at 10^3 KHz is shown in Table (2).

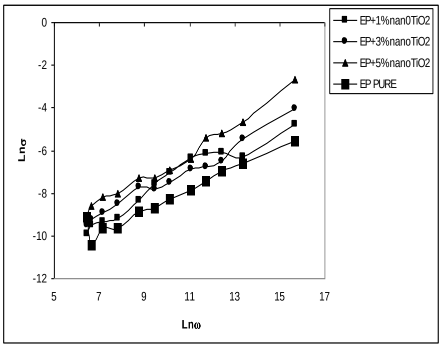
Table (2): The conductivity values for EP and EP/TiO_2composites (nano and micro)

Samples	**Ln σ at 10^3 KHz**
EP PURE	**-5.55**
EP+1% micro T iO_2	**-3.27**
EP+3% micro T iO_2	**-2.68**
EP+5% micro T iO_2	**-1.71**
EP+1% nano T iO_2	**-4.76**
EP+3% nano T iO_2	**-4.04**
EP+5% nano T iO_2	**-2.69**

Hence, it is proposed that two factors influence σ_t (ω), which are ions motions and polymer backbone (main chain) motion. Furthermore ions motion is contributed at high frequency [9,10]. The increasing of σ_t (ω), at low frequencies over (100-400) Hz is the attributed to the interfacial polarization, since the direct conductivity (σ_d.c) is significant at this region [11]. The rapidly increasing of σ_t (ω) 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.



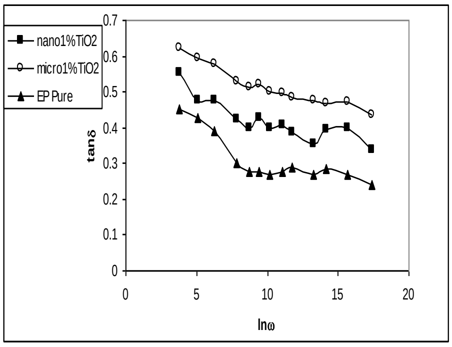
(a)



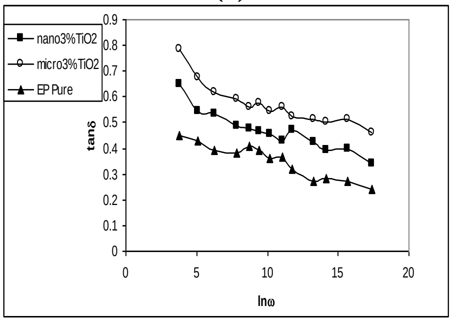
(b)

Fig (2-a,b) : Frequency dependence σ for epoxy/TiO_2 (nano and micro)of (1%,3%,5%)TiO2.

Figure (3) shows the changes of the dissipation factor tanδ with frequency, at room temperature .This change must be to interfacial polarization at the electrodes. The dissipation factor tanδ (is the degree of dielectric loss). In general, dissipation factor values for composites with higher addition of TiO_2 increasing with increasing weight percentage of TiO_2 were smaller than those with micro particle this may be was attributed to interfacial polarization. Also, the dissipation factor as can be seen from the figures increases with the decreasing in frequency due to the interface polarization. At low signal frequency range, when the electrical dipoles are able to follow the variation of the electric field, the dissipation factor decreases with the increases frequency, and composites with micro TiO_2 have higher values than that with nano particles TiO_2 [12,1.3].



(a)



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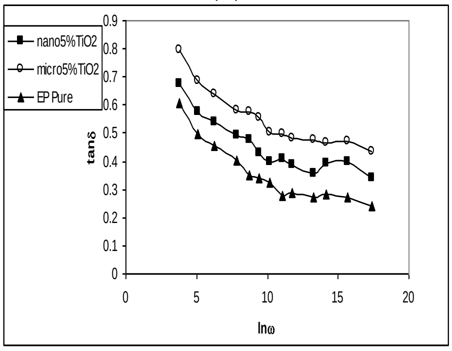

(c)

Fig (3-a, b, c): Frequency dependence of tanδ for EP Pure and EP/TiO_2 (nano and micro composites).

Conclusion

1-Dielectric constant and dissipation factor values for all composites decrease with increasing frequency.

2-Dielectric constant values for epoxy composites with micro sized particles of TiO_2 have the highest (5%) values 18.3.

Reference

[1]. P. Chahal, R. P.Tummala, M.G.Allen, and M.Swaminathan, IEEE *"Transactions on components packagng and Manufacturing Technology'*, part B: Advanced packing (**1998),**21,1840.

[2] C.J.Dias, R.Marat , P.Inanico, and N.Mendes, IEEE,*Transaction on Dielectric and Electrical insulation* , (2004),11,35.

[3]. N.S.Bhavanesh, and J. Gopalakrishnan, *J. of Mater.Chem*.(1997),7,2297.

[4]. A.L. Linsebigler, G.Lu and J.T Yates , *Chem. Rev* (1995).95,735.

5-G.S. Oehrlein *J.Appl.Phys*. **(1986),**59,1587.

[6]. D.Hull and T.W.Clyne,"*An interaction to composite material", Oxford University*, press, Oxford, U.K.(1994).

[7]. T.J.Lewis, IEEE *Transation on Dielectric and Electrical insulation* **,(1994),**Vol.1,812.

[8]. T.J.Lenis, *J.of Phys. D:Appl. Phys*.(2005),38,202.

[9]. L.Cheng, L.Zheng, G.L.Yao. and Q.Y.Jing, *J.Nanoparticales* **,(2008),**1.

[10]. H. Janssen,J.M.Seifert and H.C.Karner, *Dielectricand Electrical insulation,Transactions on Dielectric and Electric Insulator***,(1999),**6,5.

[11]. G.C.Psarras,E.Manolakaki, and G.M. Tsangaris" *Dielectric dispersion and ac conductivity in—Iron particles loaded—polymer composites",* Composites Part A: Applied Science and Manufacturing **(2003),** 34, 1187-1198.

[12]. M. Al-Haj Abdallah, Y. Alramadin ,M. Ahmad, A. Zihlif, S. Jawad, and A. Alnajjar *"Electrical Characterization of Metal Fiber-Polyester Composite"* International Journal of Polymeric Materials **(1997),** 37 , 33 - 42.

[13]. A.R. Donnelly, and K.P. Varlow, " *Nonlinear dc and ac conductivity in electrically insulating composites"* Dielectrics and Electrical Insulati