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Dielectric properties of nanosilica filled epoxy nanocomposites

M G VEENA[1], N M RENUKAPPA[1,*], KUNIGAL N SHIVAKUMAR[2] and S SEETHARAMU[3]

[1]Department of Electronics and Communication Engineering, Sri Jayachamarajendra College of Engineering, Mysuru 570006, India
[2]Center for Composite Materials Research, Department of Mechanical Engineering, NC A&T State University, Greensboro, NC 27411, USA
[3]Materials Technology Division, Central Power Research Institute, Bengaluru 560080, India
e-mail: veenamaheshn@gmail.com; renunijagal@gmail.com; kunigal@ncat.edu; sub.ramu@gmail.com

Abstract. This paper presents the development of epoxy-silica nanocomposites and characterized for dielectric properties. The effect of nanosilica loading (0–20 wt%), frequency, temperature and sea water aging on these properties was studied. Transmission electron microscopy (TEM) analysis of the samples showed an excellent dispersion. However, at higher silica loading TEM showed inter-contactity of the particles. The dielectric constant (ε') increased with silica loading and reached an optimum at about 10 wt%. The ε' of the nanocomposites showed linear decrease with frequency whereas AC conductivity (σ_{ac}) increases. The σ_{ac} and ε' increased marginally with temperature and sea water aging.

1. Introduction

Polymer nanocomposites are the 21st century engineering materials with wide range of dielectrics applications, i.e. applications that make use of electrically insulating or nearly insulating behavior. This is because of the need of the electronic industry for dielectric materials in electrical insulation, encapsulation, substrates, interlayer dielectrics in a multilayer ceramic chip carrier, printed circuit boards and capacitors; and because of the rising importance of smart structures which use dielectric materials for piezo-electric, ferroelectric and pyroelectric devices that provide sensing and actuation, etc. Nanoparticles tend to affect polymer resins differently and could change the material properties compared with conventional microfiller filled polymer composites. Several research groups worldwide have now been able to document some significant improvements that can be made in the electrical, thermal and mechanical properties of polymer composites through the incorporation of nanoparticulates [1–4]. The inclusion of nanosilica particles into epoxy polymer is currently one of the most common ways to make polymer nanocomposites. The research interests in epoxy-silica nanocomposites can be largely divided into studies on the mechanical, thermal [3, 4] and dielectric properties [1, 5, 6]. Investigation on the dielectric properties of epoxy-TiO₂ with 10% TiO₂ micro and nanoparticles and found that dielectric constant improved with microparticle loading and nanoparticle addition decreases the dielectric constant [1]. Increase in interfacial region in nanocomposites created a zone of altered polymer properties which reduces the dielectric constant of nanocomposites [6]. The dielectric properties of nanosilica filled epoxy showed different results where dielectric constant notably increased, while the dielectric losses were lowered [5]. The investigation on the dielectric properties and ionic conductivity of epoxy-silica micro/nanocomposites increased above the glass transition temperature significantly [7]. AC conductivity for epoxy-TiO₂ composites as a function of frequency, at the high temperature range has been investigated and concluded that, at high frequencies the ac conductivity follows universal power law, weakly depends on temperature at low frequency region, and in the intermediate region dipolar relaxation processes are present, which depends on the intensity of filler content [8]. It is very important to know the variation of dielectric properties close to service conditions as a result of absorption of moisture by polymer micro/nanocomposites in practical applications. As the duration of moisture immersion increases, the ε' also increases due to the fact that water which has a high dielectric constant, enters the bulk of the system. This increase in dielectric properties is not desirable for insulating materials for performance at high voltages [9]. Further, water shell model has been developed, describing the dielectric behavior of epoxy-silica micro/nanocomposites at low frequencies [10].

*For correspondence

In most of the above studies, nanocomposites were subjected to lower frequency range and temperature. However, an investigation on the effect of high frequency range and sea water aging on dielectric properties for nanocomposites is very limited. In this paper, SiO_2 nanoparticles were incorporated into an epoxy with different nanoparticle concentration. Variation of dielectric properties and AC conductivity with temperature and exposure to sea water environment were investigated.

2. Experimental

2.1 *Materials*

The epoxy resin SC 79 (SC 79 Part-A, Diglycidylether of bisphenol A (DGEBA)) was used to fabricate nanocomposites with epoxy equivalent weight of 162 g/mol. We have used cycloaliphatic amine (SC 79 Part-B) with an amine hydrogen equivalent weight of 64.8 g/mol as the hardener/curing agent. Both the resin and curing agent were supplied by Applied Poleramic Inc. The density of the resin (SC 79 part-A) and the curing agent (SC 79 part-B) is 1.17 g/cm^3 and 0.95 g/cm^3, respectively. The viscosity at 23°C is 320 cps which increases to 950 cps after 7 h. Nanopox F400 from Nanoresins AG, Germany with epoxy equivalent weight of 295 g/mol nanofiller was used in this study. It has a concentration of 40 wt% of silica nanoparticles in DGEBA epoxy resin. This product was supplied as colloidal silica sol in epoxy resin prepared using a sol–gel process. Calculated values of Nanopox (according to the data sheet of the supplier) to have 5, 10, 15 and 20 wt% of silica were used in our nanocomposite samples.

2.2 *Nanocomposite fabrication*

The processing flowchart of epoxy-silica nanocomposites preparation is shown in figure 1. The required quantity of epoxy resin and Nanopox F400 were measured and pre-heated separately at 60°C for 2 h. The mixture was gently stirred for 15 min through mechanical stirrer ($\sim$1000 RPM) by adding SC 79B. The mixture was then degassed in vacuum oven under a pressure of 762 mmHg to eliminate the entrapped air molecules (bubbles). Then the composite

was molded into a flat plate and cured in the oven. The curing temperature was increased with a ramp rate of 5°C/min to attain 60°C and vitrified for 4 h, thereafter it was decreased to room temperature with a ramp rate of 1°C/min. The panels were then post cured with a ramp rate of 2.5°C/min till 60°C temperature was attained. Then, ramp rate was changed to 1°C/3 min till a temperature of 90°C was attained, thereafter it was changed to 1°C/min upto 120°C and remained at this temperature constant for 1 h. Finally, the temperature was decreased to room temperature with a ramp rate of 1°C/min. Using the above-mentioned process, both neat and nanosilica modified epoxy nanocomposite panels were fabricated.

2.3 *Transmission electron microscopy (TEM)*

Transmission electron microscopy (TEM, JEOL 100 CXII) with standard magnification of 360x to 320kx was used to analyze the dispersion of SiO_2 nanoparticles in the epoxy matrix at an acceleration voltage of 100 kV. TEM specimens were trimmed using an ultramicrotome (Ultracut-1, UK) with a diamond knife, and the section specimens (ca. 70 nm in thickness) were placed in 200 mesh copper grids for observation.

2.4 *Dielectric properties*

The dielectric properties of the silica filled epoxy nanocomposites in the present study are measured using standard ASTM test procedures. Nanocomposite samples were prepared at least 24 h prior to the experiment and they are kept under vacuum evacuation before they are used for the measurements. In the present study the effect of dielectric properties is studied in the frequency range from 10 mHz to 30 MHz. The capacitance (C), tanδ and conductance (G) of the prepared samples were measured over the high frequency of 1 KHz to 30 MHz at varying temperature from 25 to 150°C using HP 4281A precision LCR meter. The low frequency measurement from 10 mHz to 1 KHz of dielectric constant, dissipation factor, is measured using Frequency Domain Spectroscopy (FDS)/Polarization and Depolarization Current (PDC) analyzer (DIRANA, Model EN 50110-1). In addition, the prepared
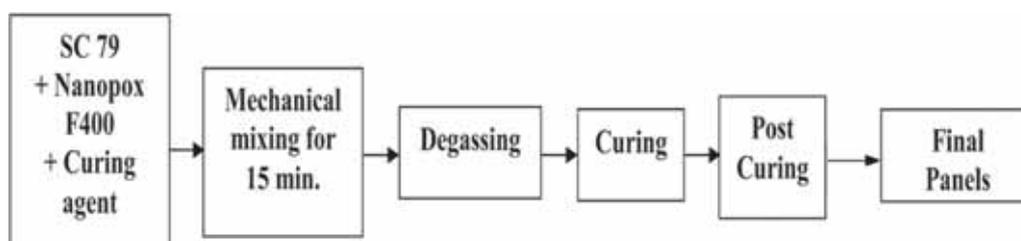

Figure 1. Flowchart for processing of epoxy-silica nanocomposites.

nanocomposite samples are immersed in sea water for 96 h and repeated the experiments at room temperature.

From the measured value of capacitance and conductance, the dielectric constant (ε') and AC conductivity (σ_{ac}) of the specimen were calculated from Eqs. (1) and (2).

$$\varepsilon' = \frac{C \times t}{\varepsilon_0 \times A} \qquad (1)$$

$$\sigma_{ac} = \frac{G \times t}{A}, \qquad (2)$$

where C is the capacitance in farads, ε_0 is the dielectric constant (8.854×10^{-12} F/m) in free space, t is the thickness of the specimen in cm, A is the area of the electrode in cm^2, and G is the conductance in Siemens.

2.5 *Sea water aging*

Sea water aging was carried out by exposing the epoxy nanocomposite specimens in a sea water bath at 25°C and at a relative humidity of 95% with a laboratory humidity chamber manufactured by M/s C.M. Equipment and Instruments (India) Pvt. Ltd., Bengaluru, India. The aging of the samples was done for 24, 48, 72 and 96 h. After each duration of aging, the samples were removed from the sea water bath and tissue paper is used to remove water on the surface and then the electrical measurements were carried out.

3. Results and discussion

3.1 *Dispersion and morphology*

Assessment of the nanoparticle dispersion and morphology can be understood from the TEM pictures of the composites. Figure 2 shows the TEM images of neat epoxy and silica filled epoxy nanocomposites. All the images were magnified to 100 kX. These images show agglomerate-free uniform dispersion of silica nanoparticles in epoxy up to 10 wt% of loading. For 20 wt% loading we observe agglomerates of silica particles. So, it can be expected that at this loading, the electrical properties will be low compared to lower loading because agglomeration indicates filler–filler interaction and less filler-epoxy matrix interaction. The size varied from 11 to 44 nm. The size of the majority of the nanoparticles was in the range of 14–21 nm which agrees with the supplier's information.

3.2 *Effects of frequency, filler loading and temperature on dielectric constant*

The dielectric constant (ε') variations of epoxy-silica nanocomposites with respect to frequency from 10 mHz to 30 MHz is as shown in figure 3a. There is a marginal decrease in the ε' over the measured frequency range irrespective of the filler loading was observed. The ε' increased with silica loading and reached an optimum at about 10 wt%. Further, the ε' decreased with increase in filler loading. The ε' decreases with increase in frequency from 10 mHz to 30 MHz. The rate of decrease is larger in the frequency region from 10 mHz to 0.1 Hz indicating "low frequency dispersion" (LFD) or "quasi-DC (QDC) behavior [10, 11], and a marginal reduction is observed over 0.1 Hz to 100 KHz. The variation of ε' is low above 100 KHz. Dielectric constant is a frequency dependent parameter in polymer composite systems. From figure 3a, as the electric field frequency increases, the bigger dipolar groups find it difficult to orient at the same pace as the alternating field, so the contributions of these dipolar groups to the ε' goes on reducing resulting in a continuously decreasing with increase of nanosilica into epoxy. Much work has been made for ε' study [12–14]. Dielectric constant usually increases if polymers are filled with inorganic fillers of micrometer size by several tens wt%. Conversely, the dielectric constant is found to decrease in many cases, if polymers are filled with fillers of nanometer size by several wt%. However, in the present work, the ε' was found to increase with increase in nanoparticle content (0–10 wt%) into an epoxy resin and 10 wt% has been considered as optimal value of filler loading.

In the epoxy-silica nanocomposites, ε' depends on the individual dielectric constant of epoxy and the fillers as well as the filler loading. The ε' associated with the epoxy component in the nanocomposite will still exist in spite of the polymer chain immobilizations. This is because the nanocomposite will have several dipolar groups which are not interacting with the nanoparticles and so they would be free to orient with the applied electric field. An increase in the nanofiller concentration in epoxy results in an increase in number of nanoparticles and with the individual dielectric constants of each of these particles contributing to increase in the ε'. The decrease in dielectric constant above 10 wt% of nanofiller loading is due to the thickness of less stoichiometrically cross-linked layer (specific gravity interlayer of several nanometers) arising from high surface energy of nanofillers and/or other species. It is assumed that this inter layer thickness may not be comparatively thick as compared to the other layers, which prevents the contact between the filler and polymer matrix, giving rise to reasonable contribution from this layer to free volume and reduction of dielectric constant. This observation has been explained by the multi-core model [15]. The other possible reasons in the reduction of dielectric constant may be due to several factors, like the restriction of polarization mechanisms in the bulk of the material. Secondly, the filler density will also determine the number of nanoparticles in the material bulk.

From figure 3b, it is observed for epoxy-silica nanocomposites the dielectric constant slightly increasing over the temperature range 25–150°C. This may be due to,

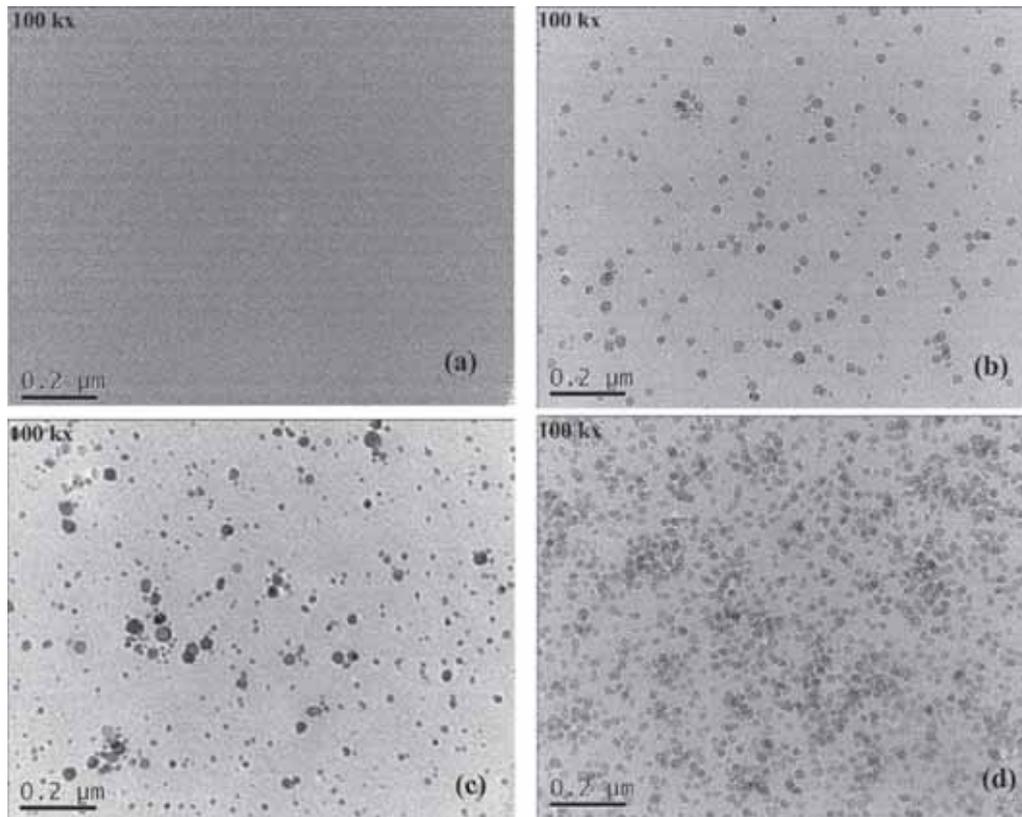**Figure 2.** Transmission electron microscope images of epoxy-silica nanocomposites: (**a**) neat epoxy, (**b**) 5 wt%, (**c**) 10 wt% and (**d**) 20 wt%.

for a glassy state epoxy the molecules of cured epoxy lose their long-range segmental mobility and the molecular motions are hindered by the cross-linking nature of epoxy network. Therefore, the effect of dipolar relaxation due to long-range molecular mobility does not dominate the material dielectric behavior. However, there exists local relaxation processes associated with the dipolar orientation of the side groups pending to the main polymer chain contributing to the dielectric properties. At higher temperature dielectric relaxation due to the long-range segmental mobility becomes obvious and shows increase in ε' [7]. In the present study, such behavior was not observed and hence effect of temperature is not much significant in epoxy-silica nanocomposites.

3.3 *Effects of frequency, filler loading and temperature on* tanδ

From figure 4a, it was noted that tanδ decreases with increase in frequency, tanδ peak appearing between 0.01 Hz to 1 Hz and between 75 KHz to 30 MHz slight increase in tanδ has been observed. There is a marginal increase in tanδ with 5 wt% silica loading, further, increase in silica content (>5 wt%) results in reduction in tanδ. In general, the introduction of inorganic fillers into the

polymer matrix increases the dissipation factor of the nanocomposites, since there is increase in number of charge carriers in the system. In nanocomposites, the huge area of filler polymer interfaces and the polymer chain entanglements might hinder the movement of charges in the material and consequently, can cause a reduction of the electrical conductivity and tanδ component. Therefore in the present study, decrease in tanδ with increasing frequency may be attributed to decrease in electrical conductivity due to polymer chain entanglements with large area of nanofiller interface within the epoxy nanocomposite system.

Measurement at low frequencies results in a peak in the frequency range between 0.01 Hz and 1 Hz. This may be due to the polarizations associated with the filler particles as well as space-charge polarizations at the nanocomposite interfaces and influence of electrode polarizations [16].

With increasing the filler loading, the tanδ increases due to the introduction of inorganic fillers to a polymer matrix, as there is an enhancement in the sources of charge carriers in the system. The number of nanoparticles causes an increase in the electrical conductivity with filler loading, which in turn influences the tanδ behavior. The number of nanoparticles is more, with increasing nanofiller loading, hence the inter-particle distances are less. With this effect, the electrical conduction in the fraction of first nanolayer
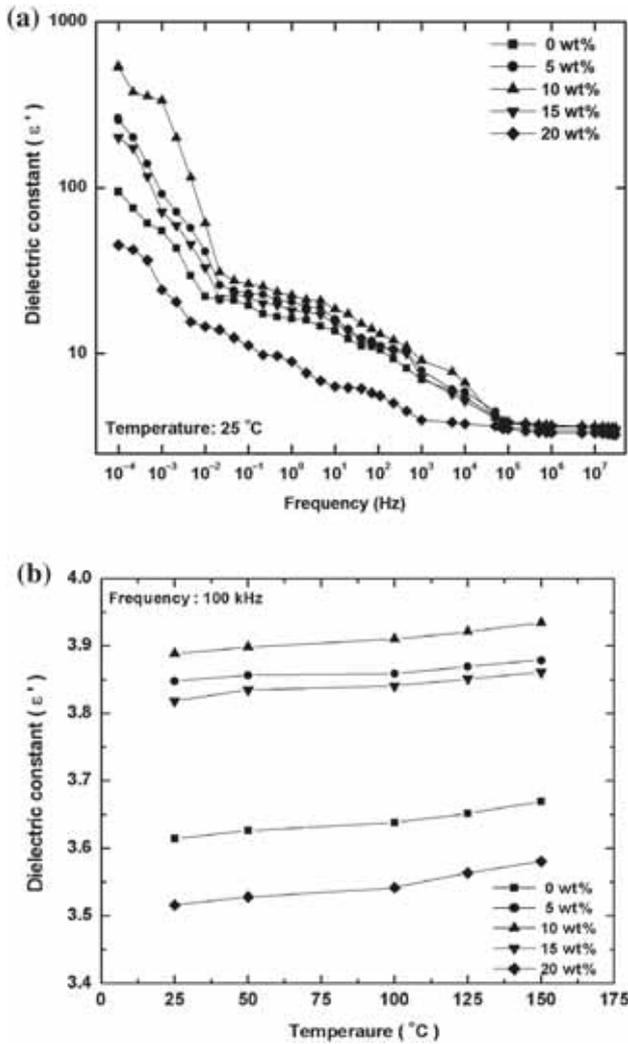
Figure 3. (**a**) Effect of frequency on dielectric constant for epoxy-silica nanocomposites. (**b**) Effect of temperature on dielectric constant for epoxy-silica nanocomposites.

Figure 4. (**a**) Effect of frequency on tanδ for epoxy-silica nanocomposites. (**b**) Effect of temperature on tanδ for epoxy-silica nanocomposites.

region is enhanced and in the fraction of the second nanolayer is lower due to the formation of overlapping regions. Therefore, the electrical conduction can take place at the close proximity of the interface regions due to the availability of free charge carriers from fillers and free charge carrier contribution from the second nanolayer is less. Therefore, there might be a reduction in the possibility of defects and charge trapping sites at the interface region. Under these circumstances, the values of tanδ in the nanocomposites will be enhanced as compared to neat epoxy [16].

The decrease in tanδ value with nanofiller loading may be ascribed to the interface barriers and chain entanglements towards the electrical conductivity, in turn the frequency dependent conductivity is sensitive to the motion of charged species and dipoles of polymers seems to be very complicated and requires further investigation. From figure 4b, it is evident that tanδ increases marginally, with

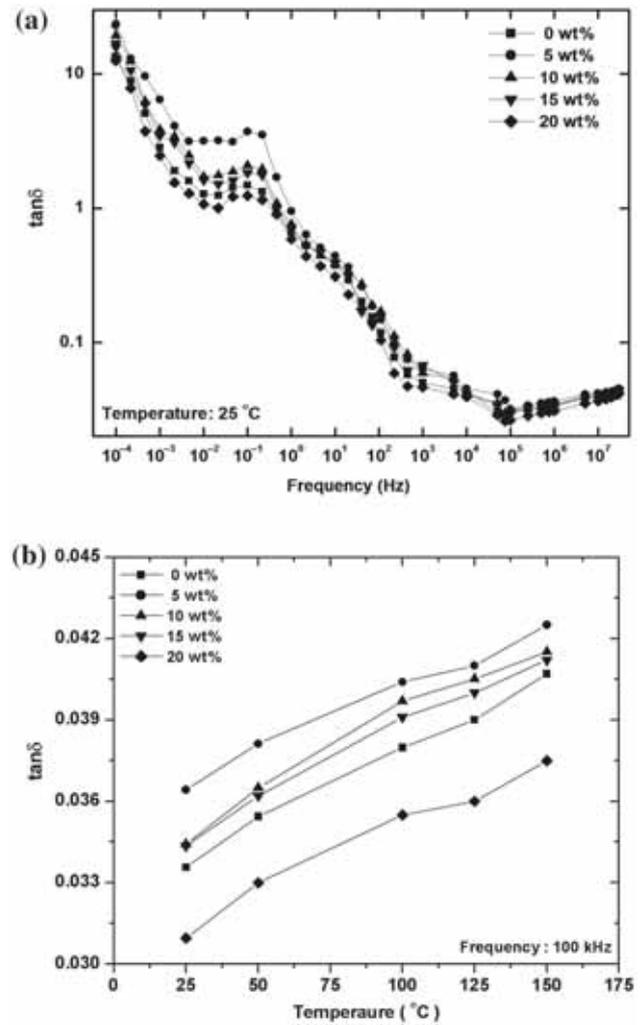increase in temperature for epoxy and epoxy nanocomposite system. The increase in tanδ with temperature is due to the thermal motions of small side-groups of polymer may be dominant, thereby increasing the tanδ. In addition, as the electrical conductivity is enhanced due to the thermally activated ions, give rise to higher tanδ.

3.4 *Effects of frequency, filler loading and temperature on AC conductivity*

The dependence of AC conductivity (σ_{ac}) with frequency is shown in figure 5a. As the effect of AC conductivity is very low below 75 KHz, the σ_{ac} effect has been studied above 75 KHz to 30 MHz. The inset graph shows the variation of σ_{ac} with SiO_2 filler loading from 10^5 to 10^6 Hz. From the figure it is observed that with increasing frequency, the σ_{ac} of epoxy and epoxy nanocomposite increases exponentially. The increase in σ_{ac} with frequency may be attributed

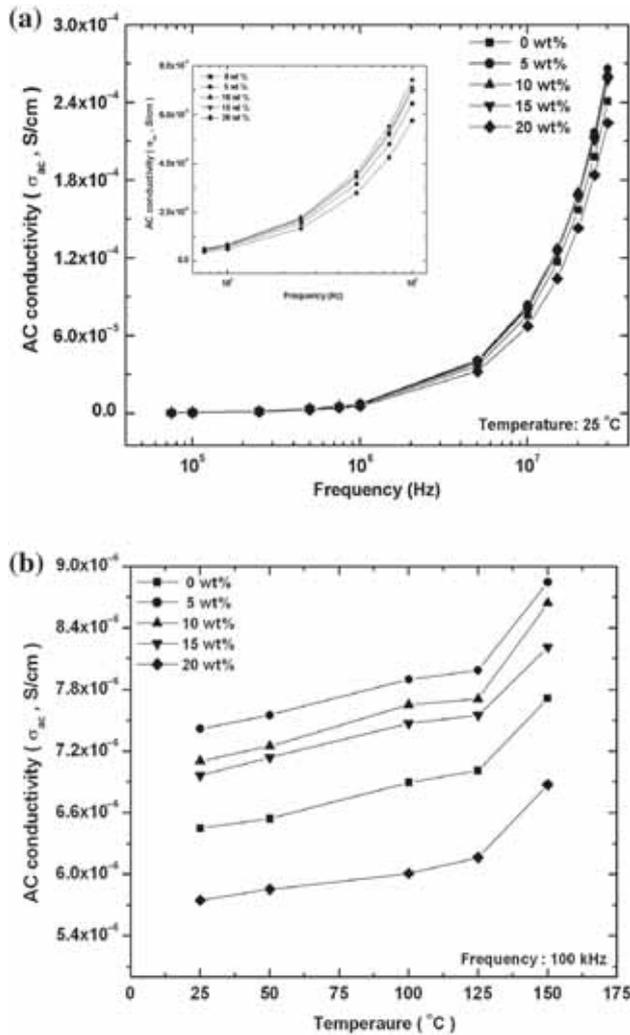**Figure 5.** (**a**) Effect of frequency on AC conductivity for epoxy-silica nanocomposites. (**b**) Effect of temperature on AC conductivity for epoxy-silica nanocomposites.

to the hopping conduction mechanism, generally consistent with the existence of a high density of states in the materials having a band gap similar to that of a semiconductor. Due to localization of charge carriers in the bulk of material, formation of polarons takes place and the hopping conduction may occur between the nearest neighboring sites increasing drastically σ_{ac} at high frequencies.

The effect of SiO_2 nanofiller loading on epoxy, σ_{ac} increased at 5 wt% filler loading. The possible reason may be, it is generally assumed that ionic conductivity is dominant in epoxy resin. The amine-cured samples, in the present study, may have hydrophilic –OH bonds that should attract impurities such as water, ions or substances that are easily dissociated into ions would be introduced into the resin of the amine-cured samples. Moreover, the Columbic attractive force between the two charge carriers with opposite polarities, which holds mobile ions in the vicinity of their parent atoms, is inversely proportional to the ε'.

Ions can, therefore, be released easily in a high ε' material and they move easily in the bulk. Since, the amine-cured samples have high ε', dissociation and transport of ions occur easily, which results in their high electrical conductivities [17].

Figure 5b shows an increase in AC conductivity upon increase in temperature. The rise of conductivity upon increasing the temperature is a common respond for polymeric samples, which may be due to the tremendous increase of the mobility of charge carriers in the composite specimen.

3.5 *Effect of sea water aging on nanocomposites*

The weight gain of the samples after the sea water treatment is listed in table 1. From table 1, it is observed that epoxy-silica nanocomposites absorb less water as compared to epoxy except for 20 wt% filler loading. This may be due to the presence of the nanoparticles increase the path length for diffusion of water molecules in volume of the epoxy matrix [18]. This shows that the incorporation of nanoparticles and its concentration do not influence the basic water diffusion mechanism in epoxy and it depends on duration of water treatment. The reduction in weight gain at 20 wt% filler loading may be due to the introduction of torturous-paths by silica nanoparticles for diffusion of water molecules [19] in the system. It has been postulated that absorption of water in the polymer is essentially Fickian and this leads to plasticization effects. Moreover, the equilibrium water content depends not only on the free volume as in the matrix, but also on the concentration of hydrogen bonds formed between water and network polar groups [20] and the interfacial effects.

3.6 *Effect of sea water aging on dielectric properties and AC conductivity*

From figure 6, the dielectric constant, tanδ and AC conductivity increased linearly with sea water aging (96 h) of epoxy and epoxy nanocomposites is observed. From figure 6a, the increase in dielectric constant may be ascribed to the stronger dipoles of water compared to the matrix system and the nanocomposites. Besides, the absorbed water molecules may

Table 1. Weight gain of the samples after the sea water treatment.

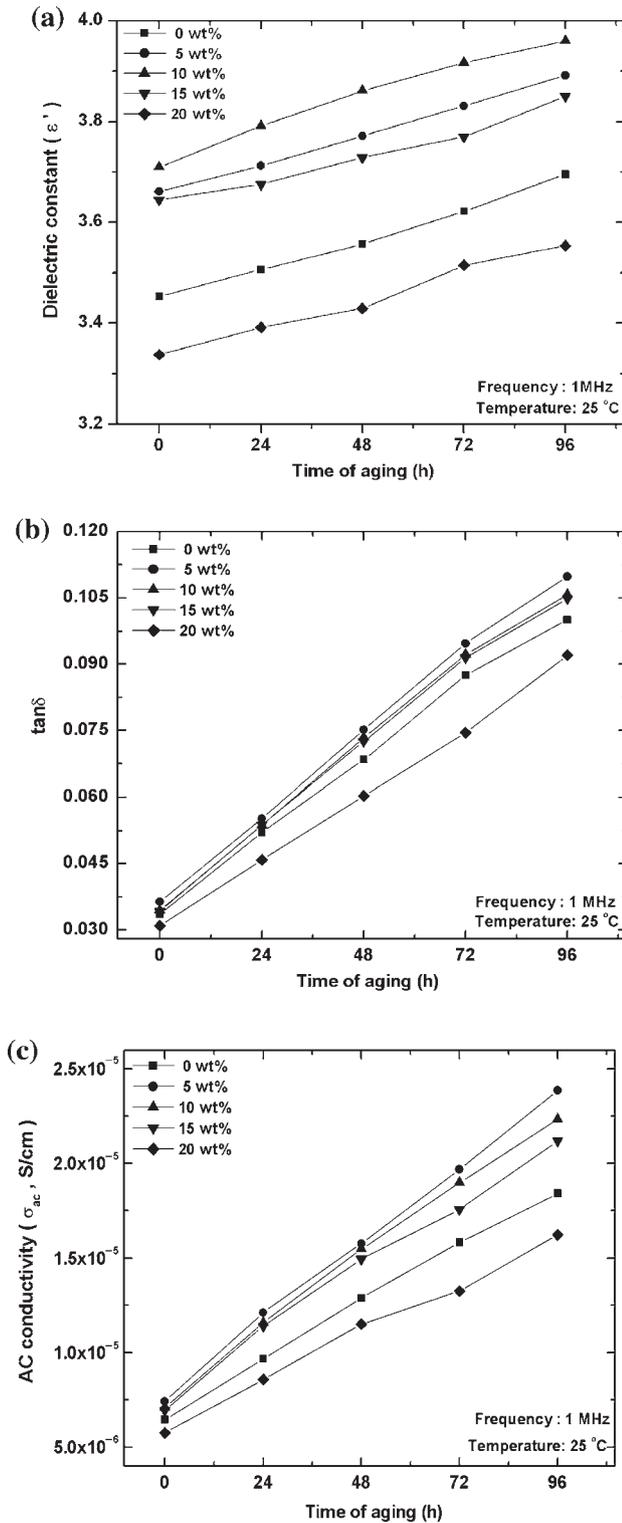| Duration (h)	Filler loading (wt%)				
	0	5	10	15	20
24	0.28671	0.12354	0.20982	0.19767	0.28755
48	0.30891	0.19698	0.23234	0.22026	0.39561
72	0.38761	0.21765	0.31254	0.24560	0.41573
96	0.43131	0.26264	0.35414	0.28533	0.45563

Figure 6. Effect of sea water aging on epoxy-silica nanocomposites: (**a**) dielectric constant, (**b**) tanδ and (**c**) AC conductivity.

damage the polymer main chains and increase the segmental mobility by plasticization, increasing free volume or even breaking weak bonds, and thus can increase the total dipole strength of the nearby segments, increasing the dielectric constant. With increase in filler concentration, the hydrophobic surface of nanofillers increases more and more, by making less fraction of epoxy matrix, confirming in reducing the water sorption in epoxy materials [21]. The effect of sea water aging on epoxy-silica nanocomposites shows an increase in tanδ and AC conductivity value up to 96 h was observed from figures 6b and 6c. The increase in electrical conductivity of heterogeneous system influences the tanδ value. The increase in AC conductivity is mainly due to surface adhesion of the ions of sea water and as such the electrical conductivity increases. For sea water sorption, the rate of sorption is higher due to ionic nature of water. The absorbed water will not be in one layer but in different layers of the epoxy matrix and therefore the transfer of charge carriers occurs rapidly and conductivity increases. The first few water molecules may be firmly bounded to the nanoparticle. Other water molecules may loosely bound by van der Waals force.

Therefore, concentration of water in the matrix may be sufficient for conduction and is likely to provide a channel for charge carriers resulting in increased conductivity [10]. Secondly, the high polarity of the water molecules can attract charge carriers, particularly at the filler–matrix interface and become significant resulting in increased conductivity [22].

4. Conclusion

Based on the results of the present investigation:

- It can be concluded that epoxy-silica nanocomposites characterized by TEM showed non-agglomerated distribution of the nanosilica particles up to 10 wt% and agglomerate of particles at 15 and 20 wt% is observed which has effect on dielectric properties.
- The dielectric constant increased with nanosilica loading and reached an optimum at about 10 wt%.
- The effect of frequency on the epoxy-silica nanocomposites showed linear decrease in dielectric constant, on the other hand nonlinear decrease in tanδ and slight increase at higher frequencies, whereas, the AC conductivity increased drastically at high frequencies.
- The weight gain after sea water treatment for epoxy-silica nanocomposites shows less absorption of water as compared to epoxy.
- The dielectric properties and AC conductivity increased with sea water aging (96 h), due to absorption of sea water at the interfaces between the polymer and inorganic filler.

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