

Effect of Pre-treatment of Nanofillers on the Dielectric Properties of Epoxy Nanocomposites

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ABSTRACT

Early researches show that the nano filler dispersion has a significant effect on the dielectric properties of resulting epoxy nanocomposites. In this paper a comparative study on the dielectric permittivity, AC breakdown behaviours and glass transition temperature of epoxy nanocomposites loaded with both treated and untreated nano SiO₂ fillers has been carried out. The epoxy resin (LY556), commonly used in power apparatus, was used to study the dielectric behaviour of epoxy. Silane was used as a coupling agent to pre-treat the nano particles. The results indicate that the surface treatment of nanoparticles has a strong effect on the dielectric properties of resulting nanocomposites as surface treated nanoparticles with better dispersions is believed to hinder the polymer chain mobility and charge transportation through the interface region between epoxy and nanoparticles.

Index Terms - Nanocomposites, epoxy resin, nanofillers, dielectric properties, dielectric strength, silane, surface treatment.

1 INTRODUCTION

EPOXY resin is one of the most commonly used thermosetting materials in high voltage apparatus as insulation due to its excellent mechanical, electrical properties and chemical stability. In recent years, the nano reinforced epoxy resin has attracted a wide interest as it enhances the epoxy's properties significantly [1-3]. Researches on nano reinforced epoxy resin composites have been carried out ceaselessly. The previous investigation has shown that the epoxy/nanocomposites demonstrate some advantages in both mechanical and dielectric properties [4, 5] compared with pure resin system and epoxy with micrometer-size fillers at a lower loading concentration (1-10 wt%). Recent researches also indicated that the interface area between nano size fillers and the base polymer materials is one of the determining factors for the dielectric properties of the nanocomposites. It has been shown that such poor filler dispersion has a negative effect on the performance of composites [22]. To improve the nano size filler dispersion, the surface treatment method has been introduced. The surface treatment can reduce the nano size fillers' surface tension and therefore result in better filler dispersion in polymer matrix. Several studies have shown

that the polymer nanocomposites with surface treated nano size fillers tend to have better insulation properties compared with those without surface treatment [23-27]. Tagami et al. studied the effects of both cure agent and dispersion method on the dielectric properties of epoxy/clay nanocomposites. The results indicate that the curing agent has a much stronger effect on the dielectric properties as compared to the dispersion method [23]. Tanaka et al. studied the effects of different modification methods on the partial discharge aging of epoxy nanocomposite with 5 wt % additions of clay. They suggest that nano particle with interaction zone effect is a possible mechanism for the improvement in PD resistance [26].

Up to now, there is still little amount of works on the properties of the nano-size particles and the dielectric properties of the epoxy composites with various of filler loading concentrations. In this paper, the authors study the effects of SiO₂ nano particles with and without surface treatment on the glass transition temperature, dielectric permittivity and break down behaviors of epoxy nanocomposites. The effect of silane used as a modification agent on the dielectric properties of epoxy nanocomposites is also discussed in detail as respect to the nanoparticle filler loading concentration. Based on the dual layer model, the possible mechanism of surface modification on the dielectric properties of epoxy nanocomposites is also discussed.

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2 EXPERIMENTAL DETAILS

2.1 MATERIALS

The materials used in this study to produce epoxy resin specimen were Araldite LY556 (Bisphenol-A type) and the hardener HY917 (anhydride type hardener), supplied by Huntsman. The epoxy resin system is widely used in practice and has an advantage of low viscosity at room temperature. Both nanoparticles of silicon dioxide (average diameter 25 nm) supplied by NanoAmor were used as fillers.

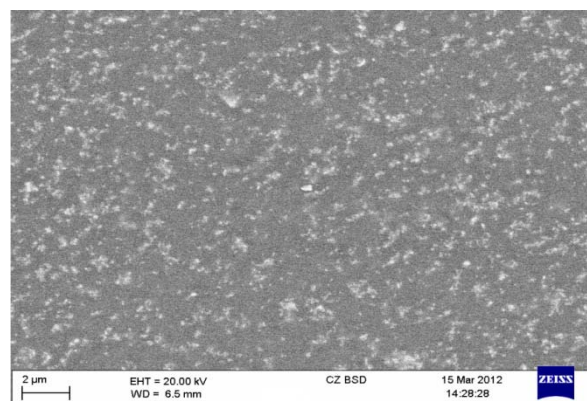
2.2 SAMPLE PREPARATION PROCESS

In this study, the epoxy resin specimens (200 μm for dielectric spectroscopy and 100 μm for dielectric strength) with loading concentrations of nano SiO_2 fillers (with and without pre-treatment) 1 wt%, 3 wt% and 5 wt% were produced. Pure epoxy resin specimens and specimens with micro-size SiO_2 fillers were also prepared for comparison purpose. The mix ratio used in this study for araldite and hardener is 1: 0.9 (by weight), according to the material specification data sheet. The concentration of accelerator is 1 part in per 100 parts epoxy resin. Before the process of preparing epoxy resin specimens, both epoxy resin and hardener were poured into beakers separately. They need to be degassed at 40 $^\circ\text{C}$ for 2 h in vacuum to remove trapped gas bubbles and moistures, and to reduce its viscosity. The surface functionalization of nano silica filler was carried out using silane as a coupling agent. After the completion of the degassing process, the nano fillers were dispersed into epoxy resin slowly with hand stirring. The hardener was added into the resulting epoxy/nano-filler solutions and then the mixture was stirred with both a high speed mechanical mixer (at 600 rpm) and an ultrasonic liquid processor for 1 h each. The mixture was degassed in the vacuum oven for another 20 min at 40 $^\circ\text{C}$ to remove gas bubbles generated during the mixing process. After degassing, the mixture was poured into the mould. Then the mould was placed into the oven at 80 $^\circ\text{C}$ in vacuum to cure for 4h followed by 4 h post-cure at 120 $^\circ\text{C}$. Epoxy resin specimens (200 μm) were prepared and then kept under vacuum for at least 24 h before dielectric measurements.

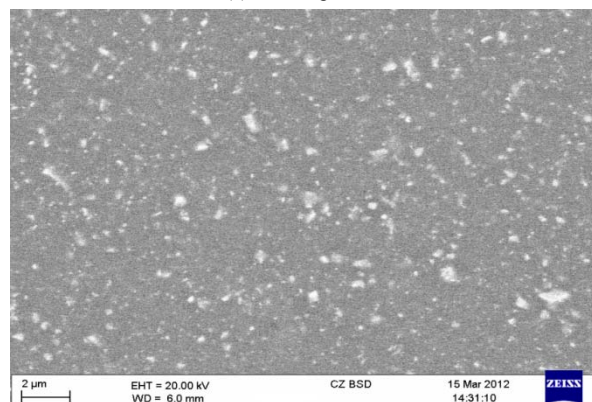
Scanning electron microscope (SEM) was used to exam the dispersion of nano-size fillers in epoxy/nanocomposites. The filler dispersion of the nano-size filler in the resin can be seen from Figure 1. The SEM results show that the treated nano fillers have better dispersion rate (dispersity of the nanoparticles in epoxy samples) compared with the untreated fillers.

2.3 DIELECTRIC SPECTROSCOPY MEASUREMENTS

The dielectric permittivity of epoxy/nanocomposites was measured over a wide range of frequencies using dielectric spectroscopy (Solatron 1260). The dielectric spectroscopy is based upon the interaction of the electric dipole moments within the sample and the applied field, and provides the dielectric properties of materials, such as relative



(a) With pre-treatment



(b) Without pre-treatment

Figure 1. SEM Image of the Epoxy– SiO_2 Nanocomposite loaded with treated and untreated nano filler.

permittivity and loss factor. By testing the sample's dielectric properties, the information about the chain mobility of polymers can be extracted. During the test, both the real and imaginary part of the dielectric permittivity of the epoxy resin composites' dielectric permittivity over a range of 1 Hz to 1 MHz were measured. The sample was cut into a circle with 25 mm in diameter and the thickness of the sample was 200 μm . The applied voltage was set to 1 V and the resulting relative permittivity and its loss characteristics ($\tan\delta$ value) were measured. During all the measurements, the temperature was maintained at 20 $^\circ\text{C}$.

2.4 DIELECTRIC BREAKDOWN MEASUREMENTS

The AC dielectric breakdown behaviours were measured by using a purpose-built kit based on ASTM standard D149-87 [6, 7]. The schematic diagram of this kit is shown in Figure 2. The sample is immersed in Dow Corning 200/20 cs silicone fluid, between two 6.3 mm steel ball bearings. The upper electrode, which the upper ball bearing is attached to, has a 50 g load added, to ensure the sample is sufficiently held in position and also, to eliminate a film of oil between the electrode and the sample, which could potentially interfere with results. The ramp testing method has been used to test the breakdown strength of epoxy samples. The process is simply applying a linearly increased voltage to the tested sample until the breakdown occurs. With this purpose-built kit, both of the breakdown voltage, V_{bd} , and time taken for breakdown to happen can

be recorded. Thus the breakdown strength, E , of the epoxy resin film can be worked out with the equation (1):

$$E = \frac{V_{bd}}{d} \quad (1)$$

Where, d is the sample thickness. During testing, a 50 Hz voltage is ramped from 0 to 18 kV at a rate of 50 V/s. The high voltage is measured using a 10000:1 potential divider and a Precision Gold WG 020 digital multi-meter.

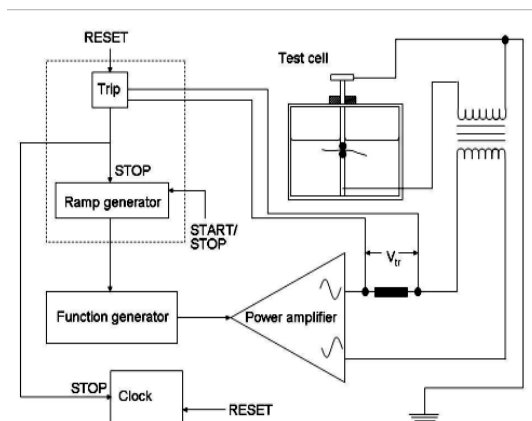


Figure 2. Schematic diagram of AC dielectric breakdown kit

The Weibull 7++ software package (Supplied by Reliasoft) has been used to analyse the results. The Weibull distribution is a continuous probability distribution that is used to describe breakdown distribution. It was firstly introduced by Weibull in 1951. It is the most commonly used probability distribution in breakdown studies due to its flexibility. It is also the IEEE recommends standard probability distribution. The Weibull distribution allows either one, two or three parameters being used to describe the breakdown distribution. In this project, two parameter Weibull is selected to analyse the breakdown results due to its best overall performance. The equation of two parameter Weibull is shown as equation (2):

$$P_f(x) = 1 - \exp\left[-\frac{(x - x_t)^\beta}{\alpha}\right] \quad (2)$$

Where $P_f(x)$ is the cumulative probability of failure at field x , x_t is a threshold field, which means that below field x_t , no failures can occur. Both α and β are known as location and shape parameters respectively.

2.5 DIFFERENTIAL SCANNING CALORIMETRY MEASUREMENTS

The glass transition temperature of unfilled epoxy resin and epoxy nanocomposites filled nano SiO_2 fillers with and without surface treatment with filler loading by 1, 3 and 5 wt% were measured by the differential scanning calorimetry (DSC) technique using a Perkin-Elmer DSC7 together with the Perkin-Elmer Pyris software. Samples with a small mass of around 5 mg had been selected and sealed in an aluminum can. The sealed can was placed in the DSC together with an empty reference aluminum can separately. During the test the temperature was increased from 40 to 170 °C with a ramp rate of 10 °C/min.

3 THERMAL CHARACTERISTIC

The glass transition temperature for epoxy resin loaded with different loading concentration has been shown in Figure 3. In addition, the effects of surface treatment on the glass transition temperature of epoxy nanocomposites have also measured. It can be seen that the presence of nano size fillers seems to reduce the T_g . Compared with pure epoxy resin, the epoxy resin loaded with 1 wt% nano SiO_2 fillers seems to have a slightly lower T_g . As the loading concentration increases, there is a further reduction on T_g .

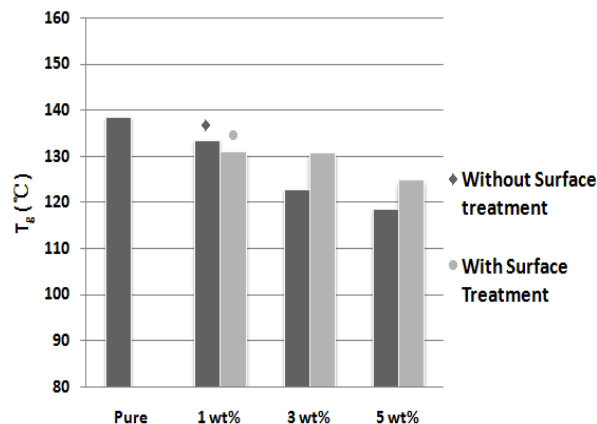


Figure 3. Glass transition temperature for epoxy resin loaded with treated and untreated nano filler.

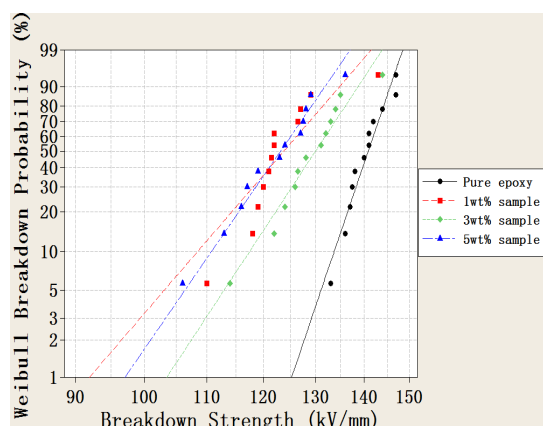
The T_g behaviour can be explained by the surface characteristic of nano filler and the interfacial region between nano filler and the polymer matrix. Early researches by Starr et al show that the interfacial region is the key factor that affects the T_g of polymer nanocomposites [8, 9]. The dual layer model proposed by Santanu and Thomas [4] can be applied to explain the behaviour of T_g observed. In this model the interfacial region around the nano fillers is classified into a tightly bound layer and a loosely bound layer. The inner tightly bound region is believed to be tightly bound between nano fillers and polymer chain by weak hydrogen bonds and therefore restrict the chain mobility. The outer loosely bound layer, on the other hand, contains polymer chains that are loosely bound. The presence of loosely bound region act as a mobile region and lead to a reduction in T_g . From T_g point of view, the two layers act in opposite senses. Reference to the early researches, a reduction of T_g has been reported when the nano filler is introduced [10], where T_g increase has also been observed in a few cases [8]. At 1 wt% loading concentration, the tightly bound region is small and the presence of a large mobile region leads to a reduction in T_g . The interfacial area between epoxy and fillers increases significantly with the increase of filler loading concentration and results in the increase of free volume due to the loosened molecular packing of the chains. Such interfacial interaction will determine the T_g [11-13]. As a result of increasing free volume, the T_g of the epoxy nanocomposites decreases as the loading concentration increases [5]. The T_g of epoxy nanocomposites loaded with treated nano fillers has a slower decrease rate with increases of loading concentration compare with the epoxy nanocomposites loaded with non-treated nano fillers. When

silane is used as a coupling agent to modify the nano fillers, tight bond can be formed between epoxy molecular and inorganic nano size fillers. Therefore, such reduction in decrease rate is believed due to the restriction of surface treated nano filler on polymer molecular chain movement which will result in the decreases of T_g .

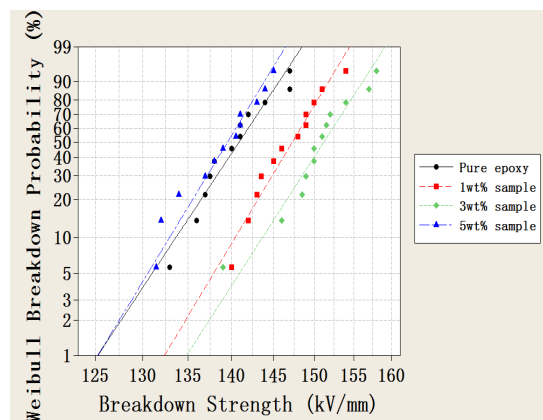
4 INSULATING PROPERTIES

4.1 DIELECTRIC BREAKDOWN STRENGTH

The AC breakdown strength of unfilled epoxy resin and epoxy nanocomposites filled nano SiO_2 fillers with and without surface treatment with filler loading by 1, 3 and 5 wt% were measured and the results were shown in Figure 4. The two parameter Weibull distribution was used to analyse the AC breakdown results, thus producing the Weibull parameters shown in Table 1.



(a) Without surface treatment



(b) With surface treatment

Figure 4. Breakdown strength of epoxy nanocomposites

The results show that there is a reduction in breakdown strength for all epoxy nanocomposites loaded with untreated fillers. The breakdown strength of epoxy nanocomposites loaded with 1 wt% untreated filler is lower compared with the breakdown strength of unfilled resin. However, when the filler loading concentration increases to 3 wt%, there is a small increase in breakdown strength compare with 1 wt% nano filled specimens. But if the filler loading concentration further increases to 5 wt%, the breakdown strength of result specimens starts to decrease

Sample	Without Surface Treatment		With Surface Treatment	
	Scale parameter (η) (kV/mm)	Shape parameter (β)	Scale parameter (η) (kV/mm)	Shape parameter (β)
Pure	140.1	39.9	140.1	39.9
1 wt%	126.0	14.3	146.9	42.7
3 wt%	130.5	19.0	151.4	43.0
5 wt%	124.3	17.5	140.5	45.6

Table 1. Weibull parameters from MLE for epoxy nanocomposites loaded with both treated and non-treated nano size filler.

again. Unlike the epoxy nanocomposites loaded with untreated nano fillers, epoxy nanocomposites loaded with treated nano fillers has better breakdown strength compare with the unfilled epoxy, as shown in Figure 4b. It can be clearly observed that the breakdown strength of epoxy resin loaded with 1 wt% treated filler is slightly higher compared with the unfilled epoxy specimens. When the loading concentration increases to 3 wt%, there is a further increase in both shape and scale parameters. However the breakdown strength starts to reduce with further increase of the filler loading concentration to 5 wt%. It has been reported by Preetha et al that a similar trend of breakdown strength behaviour has been observed in epoxy nanocomposites loaded with nano Al_2O_3 fillers [14]

The above results show that the surface treatment of nano fillers has a significant effect on the breakdown behaviour of nanocomposites. Early researches have already shown that there is an interfacial region between polymer matrix and nano fillers which is formed due to the interaction between polymer matrix and nano fillers. Thus the interfacial characteristic between polymer matrix and nano fillers seems to play an important role in the insulation performance of polymer nanocomposites. To help understanding the interface behaviours, Singha and Thomas have proposed a dual layer model [4]. In this model Singha and Thomas have classified the interfacial layer between polymer matrix and nano fillers into two layers, as shown in Figure 5. The inner layer is the tightly bound layer and the outer layer is the loose polymer layer.

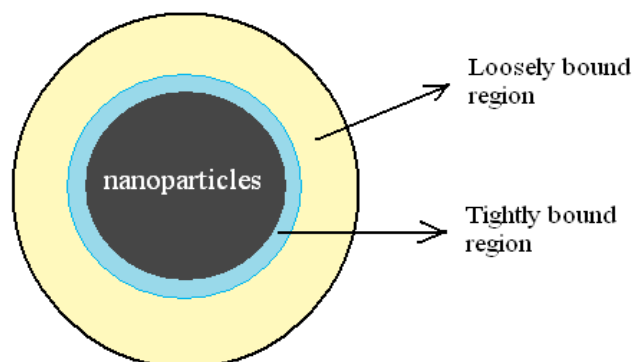


Figure 5. Dual layer model

The According to the dual layer model, the loosely bound region is more electrically conductive in nature, whereas the tightly bound region could act as charge traps to reduce the number of free charge carriers for charge transport. Thus it is clear that the inter-particle distance has a significant effects on the dielectric strength of epoxy nanocomposites. The overall reduction of the dielectric strength of epoxy nanocomposites loaded with un-treated nanoparticles is due to the agglomeration of un-treated nanoparticles could act as defects and therefore reducing the dielectric strength in epoxy nanocomposites. In addition, early researches also show that the mobile charge carriers could travel easier through the loosely bound region, so those charge carriers could travel through the bulk of polymer nanocomposites much easier under the applied field and result in lower dielectric strength. On the other hand, when charge injection takes place in the bulk of the specimens, counter ions will be adsorbed around the nano fillers as a result of electrostatic Coulomb force and other force. The presence of such electric double layer will result in an inner field at the tightly bound region. Thus when charge carriers are injected into the polymer nanocomposites, the nano fillers can scatter the charge carriers through the bulk of the specimens and result in higher breakdown strength. The combination of the effects above causes the reduction of the breakdown strength in epoxy nanocomposites.

The average inter-particle distance of epoxy nanocomposites is shown in Table 2. The average inter-particle distance and the surface area have been calculated using the equation given by Tanaka et al [15]. Higher inter-particle distance leads to a larger volume fraction of the loosely bound region, which contribute to the overall reduction of the dielectric strength of epoxy nanocomposites loaded with un-treated nanoparticles. The epoxy nanocomposite loaded with 3 wt% nanoparticles has slightly higher dielectric strength as compare to the 1 wt% samples. This is because the electric double layer effects start to play a more important role as the number of nanoparticles increase, therefore lead to a small increase in dielectric strength. A further increase in the filler loading concentration to 5 wt% leads to another reduction in breakdown strength. There is also a further decrease in inter-particle distance. The possibility for the loosely bound region (with a thickness of a few ten nm) of interface to overlap with each other is sufficiently high. Such overlapping of the loosely bound region is believed to form conductive paths that allow charge carriers travel through much easier. Thus the dielectric strength of epoxy nanocomposite might further reduce drastically.

Table 2. Inter-particle distance and surface area of nano filler in epoxy nanocomposites.

Sl. No.	wt%	Interparticle distance (nm)	Surface area (km ² /m ³)
1	1	68.4	3.41
2	3	39.5	10.31
3	5	29.3	17.35

Compared with unfilled epoxy resin specimens, the nanocomposites with 1 wt% loading of treated fillers has higher breakdown strength. Surface treated nano fillers can disperse more homogeneously in base resin. So the variance of the inter - particle distance is small in surface treated nanocomposites as compared to untreated one. Therefore, for epoxy nanocomposites loaded with treated nano fillers, the nano fillers inside the polymer can be treated as isolated particles under lower loading concentrations. Also, after surface treatment, the silane will attach to the surface of nano fillers to form chemical bounds. This will lead to a decrease of surface tension of nano fillers and therefore enhance the interfacial adhesion between epoxy resin and inorganic nano filler. Thus, it is able to reduce the voids in epoxy nanocomposites loaded with surface treated nano fillers and result in increase of breakdown strength. With 3 wt% loading concentration, the breakdown strength increases slightly as the interfacial area plays a more important role. However, further increase the filler loading concentration to 5 wt% will also cause overlapping of the loosely bound region and finally result in a reduction in breakdown strength.

4.2 DIELECTRIC PERMITTIVITY

The variations of dielectric permittivity as a function of frequency for epoxy nanocomposites with both treated and untreated SiO₂ nano fillers and with different concentrations are shown in Figure 6.

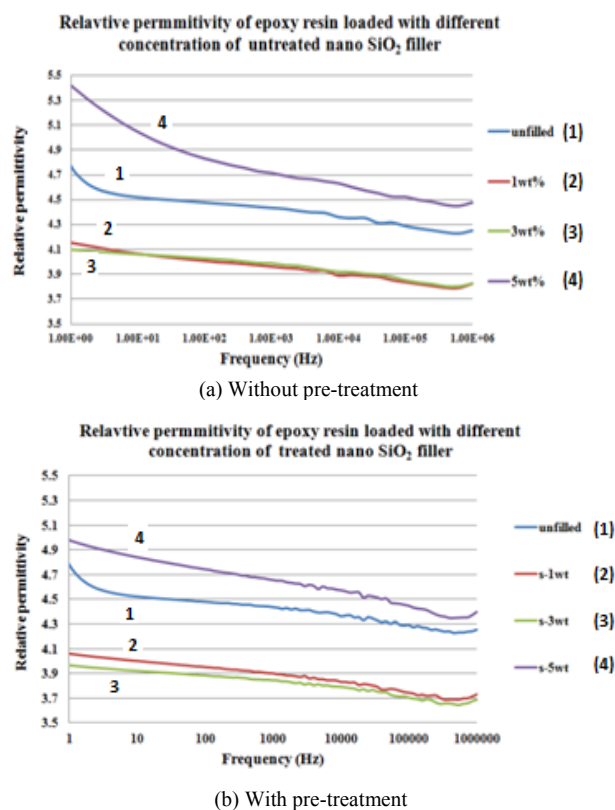


Figure 6. Variations of permittivity of epoxy-SiO₂ nanocomposites with respect to frequency.

It is observed from the measurement results that effective permittivity of epoxy nanocomposites is strongly affected by the presence of inorganic nano fillers and their

surface characteristics. It can be seen from Figure 6a that the epoxy resin nanocomposites with 1 wt% loading of untreated fillers has a lower permittivity than unfilled epoxy resin in the measured frequency range. When the filler loading concentration increases to 3 wt%, the relative permittivity is similar compare with 1 wt% nanocomposites. Further increase the filler loading concentration to 5 wt% leads to an increase in permittivity. Compared with unfilled epoxy resin, the epoxy nanocomposites loading with 5 wt% untreated SiO₂ nano fillers has a higher effective permittivity within the measured range. A similar trend of dielectric permittivity behaviour has been observed in epoxy nanocomposites loaded with treated SiO₂ nano fillers. The effective permittivity of epoxy nanocomposites loaded with treated SiO₂ nano fillers is slightly lower compared with those loaded with untreated SiO₂ nano fillers. This observation is surprising as earlier study also stated that the permittivity of epoxy/nanocomposites increases with increasing nano filler concentration in epoxy [16]. However, there are also similar observations in the literature where the nanocomposites' permittivity drops at lower concentrations and then increases again as the filler concentration increases to a higher value (more than 0.5%) [13].

It is known that the effective permittivity of the epoxy nanocomposites is found to be governed by the polarization associated with epoxy and nano SiO₂ particles and it is also

being strongly influenced by the interfacial polarization at the interface between epoxy and nano particles as well. In a typical resin system, the permittivity of epoxy is determined by the number of orientable dipoles within the system and their ability to orient under an applied electric field [17, 18]. Thus one possibility for the decrease of permittivity in epoxy nanocomposites with lower loading concentration is due to the presence of filler reducing the electrical polarization within the composites. Interfacial or space charge polarization, which is caused by the space charge accumulation near the interface between base resin system and nano size particles, will affect the effective permittivity of composites. As the interfacial polarization only occurs under lower frequency ranges (up to 10³ Hz), therefore the presence of interfacial polarization normally leads to a clear variation of permittivity with respect to the measured frequency range. Thus the effect of interfacial polarization on the decrease of permittivity in epoxy nanocomposites with lower loading concentration could be neglected as the measured results in this investigation show that the reduction of effective permittivity in lower nano loading concentration is irrespective of the frequency.

Another possibility is the reduction of mobility of the dipolar groups within the composites will reduce the polarization within the composites [19]. When the nano fillers are introduced into the polymer materials, an interaction region between polymer matrix and nano fillers is formed. The dual layer model proposed by Singha and Thomas [4] can be applied to explain the dielectric

permittivity behaviours observed and it is discussed in section 3.2. In epoxy nanocomposites loaded with 1 wt% nano fillers, the presence of tightly bound region will restrict the mobility of epoxy chain and the motion of free dipolar functional groups within the epoxy chain in the bulk material [20]. Such restriction of chain mobility hinders the dipole polarization of the polymer chain and therefore results in a decrease in dielectric permittivity. Moreover, both the tightly bound layer and loosely bound layer contribute to the decreases in local density and therefore result in lower permittivity. When the filler loading concentration increases, the volume fraction increases and leads to a further reduction in permittivity. When the silane is used as a coupling agent, the molecular movement must be restricted through OH radicals due to silane coupling. The pre-treatment of nano fillers also leads to a better particle dispersion rate in the base polymer materials, results in a contribution to the volume fraction. Therefore the epoxy nanocomposites loaded with treated nano fillers has slightly lower permittivity compared with those loaded with untreated nano fillers. Since the presence of nano size fillers tends to reduce the effective permittivity of resulting composites, the increase of permittivity in epoxy nanocomposites with higher loading concentration becomes another interesting observation. Such permittivity increase as the loading concentration increases at a higher loading concentration could be a result of the influence of the filler's inherent permittivity on resulting nanocomposites' permittivity. The permittivity of two phase dielectric satisfies the Lichtenecker-Rother mixing rule and can be written as equation (3):

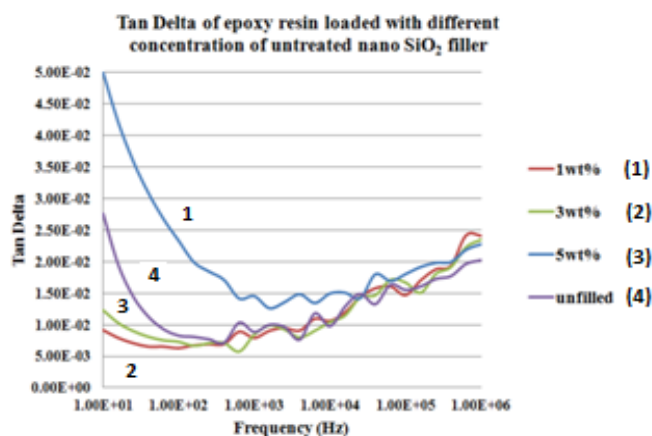
$$\log \varepsilon_c = x \log \varepsilon_1 + y \log \varepsilon_2 \quad (3)$$

Where, ε_c is the resultant composite permittivity, ε_1 and ε_2 are the permittivity of filler and epoxy and x , y are concentrations of filler and polymer. It is known that the nano size SiO₂ fillers have higher permittivity compared with the net epoxy resin. That could be one of the reasons leading to increase of effective permittivity with higher loading concentrations.

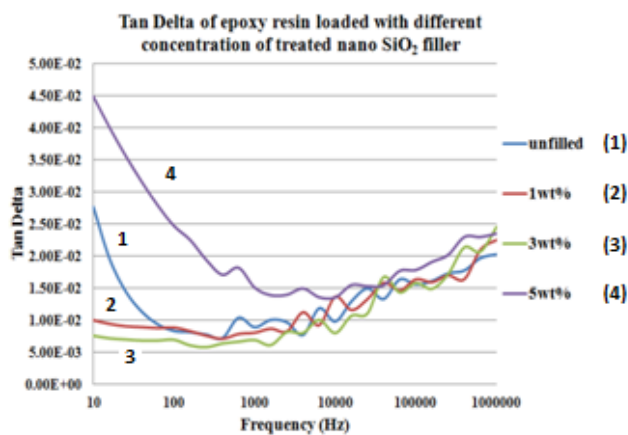
4.3 DIELECTRIC LOSS

The variation of tan delta value in epoxy nanocomposites with both treated and untreated SiO₂ nano fillers and with different concentrations are presented in Figure 7. The imaginary part of the dielectric permittivity of epoxy nanocomposites loaded with both treated and untreated nano fillers decreases with the frequency within lower frequency ranges. The imaginary part of the dielectric permittivity for both epoxy nanocomposites reaches a minimum value around 100 Hz and then begins to increase with the frequency.

It can also be observed from Figure 7 that the imaginary part of the dielectric permittivity of unfilled epoxy is higher than epoxy nanocomposites loaded with both treated and untreated nano fillers under lower frequency range (less than 100 Hz) at a lower filler loading concentration. When the frequency is higher than 100 Hz, there is no significant difference in imaginary part of the dielectric permittivity



(a) Without pre-treatment



(b) With pre-treatment

Figure 7. Variations of dielectric loss of epoxy-SiO₂ nanocomposites with respect to frequency.

between unfilled epoxy and both epoxy nanocomposites. Therefore the presence of nano size particles seems to have a stronger influence in lower frequency range whereas the dissipation factor characteristic of net epoxy resin dominates the variation within a frequency range above 100 Hz in the measurement range. Such a reduction in imaginary part of the dielectric permittivity, especially at a low frequency range, was also observed in other studies [19, 21]. As the electrical conductivity of epoxy composites contributes to its tan delta value, the variation of the imaginary part of the dielectric permittivity in both net epoxy resin and epoxy nanocomposites in lower frequency range may result from the electrical conductivity of the nanocomposites having been affected by the presence of nano size fillers. As this study was carried out under constant temperature, the influence of the relaxation time of the charge carriers on the electrical conductivity of epoxy nanocomposites can be ignored. Thus the number of charge carriers and applied frequency become dominating factor of the electrical conductivity of epoxy nanocomposites. As discussed in section 3.2, the presence of nano size fillers inside epoxy will restrict the chain mobility and result in reducing electric conductivity as such restriction limits the generation of mobile charge and the movement of charge carriers in polymer dielectrics [4, 19], especially at a lower

frequency range where the conductivity will play a more important role. Thus the variation of imaginary part of the dielectric permittivity at low frequency range may be due to the influence of inorganic fillers' electrical conductivity. Another interesting observation is that the imaginary part of the dielectric permittivity value in epoxy nanocomposites loaded with both 5 wt% treated and untreated SiO₂ nano fillers are higher than the unfilled resin. At 5 wt% filler loading concentration, the number of nano particles is more and the inter-particle distance is less compared with lower filler loading concentration, as shown in Table 2. Therefore the nano fillers are more likely to adhere to each other and the interfacial area around nano particles are likely to overlap. As a result of overlapping, the charge carries travel through the bulk of the material much easier through the overlapping region. Thus the electrical conductivity of resultant composites increases [1]. As the pre-treatment of nano particles affects the particle dispersion rate, the overlapping rate in epoxy nanocomposites loaded with treated nano fillers is less compared with epoxy nanocomposites loaded with untreated nano fillers. Thus epoxy nanocomposites loaded with treated nano fillers have lower electrical conductivity compared with nanocomposites loaded with untreated nano fillers at same loading concentration.

5 CONCLUSION

Both the dielectric permittivity and AC breakdown behaviours in epoxy nanocomposites loaded with different concentration both treated and untreated nano SiO₂ fillers have been investigated. The following conclusions may be drawn based on this study.

The presence of untreated nano size fillers shows a reduction in the breakdown strength of epoxy resin composites. Moreover, there is a significant reduction in the shape parameter (β) as well. On the other hand, the presence of treat nano fillers enhances the breakdown strength of resulting epoxy nanocomposites. The breakdown strength of epoxy nanocomposites loaded with both treated and untreated nano SiO₂ fillers appears to have a maximum value (3 wt%) with a threshold filler concentration. When the filler content is above the threshold value, the breakdown strength of nanocomposites begins to increase with filler concentration. Such reduction in breakdown strength at higher loading filler concentration is believed due to the overlapping of interfacial region between nano particles and polymer could form conductive paths that allow charge carriers travel through much easier. A reduction of permittivity has also been observed for both epoxy nanocomposites loaded with treated and untreated nano SiO₂ fillers. The reduction of permittivity at lower loading concentration is the result of a restriction of the mobility of dipole groups within the nanocomposites due to the interfacial region between epoxy and nano particles. A reduction in glass transition temperature has also been observed in epoxy nanocomposites. In summary, with the presence of nano size fillers, large interfacial region is formed between polymer and nano particles. Such interfacial region has a significant effect on the dielectric

properties of result nanocomposites. The pre-treatment of nano particles affects the particle dispersion rate in epoxy nanocomposites. Therefore the surface treatment of nano filler also affects the dielectric properties of result nanocomposites, especially at higher filler loading concentrations where the interface region is much larger compared with lower filler loading concentration.

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