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Improved Dielectric Properties of Epoxy Nano Composites

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Additional information is available at the end of the chapter

<http://dx.doi.org/10.5772/intechopen.78057>

Abstract

Epoxy-based nanodielectrics with 2, 5 and 7 wt.% of organically modified montmorillonite clay (oMMT) were prepared using high shear melt mixing technique. The interface of oMMT and epoxy of the nanodielectrics play a very important role in improving electrical, mechanical, thermal and wear properties. Therefore detailed study on the interfacial effects of filler-matrix has been investigated for understanding the chemical bonding using Fourier transform infrared spectroscopy (FTIR) and the cross linking between polymer and filler was studied using glass transition temperature (T_g) through differential scanning calorimetry (DSC). Further, positron annihilation lifetime spectroscopy (PALS) was used to determine precise and accurate value of free volume of the nanodielectrics. The interaction between the nanoparticles and polymer chains has a direct bearing on dielectric strength characteristics of the epoxy-oMMT nanocomposite system and accordingly, the ac dielectric strength of the nanodielectrics increases with the addition of oMMT into epoxy up to 5 wt.% and further increase in filler loading (7 wt.%) causes decrease in ac dielectric strength.

Keywords: Fourier transform infrared spectroscopy, positron annihilation life time spectroscopy, dielectric strength, glass transition temperature, differential scanning calorimetry, epoxy-oMMT nanocomposite

1. Introduction

The interface of surface functionalized nano-clay (oMMT) filler and polymer matrix (epoxy) of the polymer nanocomposites play a very important role in improving the electrical, thermal and mechanical properties. Therefore, detailed studies on the interfacial effects of filler-matrix on several properties have been investigated. The chemical bonding established

between epoxy and oMMT nanofiller has been investigated using Fourier transform infrared spectroscopy (FTIR). The cross linking between polymer and nanofiller was measured to determine the glassy state of the nanocomposite called glass transition temperature (T_g) by using differential scanning calorimetry (DSC). Further, the positron annihilation spectroscopy (PALS) has been utilized to determine free volume as outlined in the multi-core model [1]. Many researchers have theoretically estimated the free volume of nanocomposites and there is no experimental data on the evaluation of free volume. In the present work, PALS has been used in the accurate evaluation of free volume. A brief explanation of nanocomposite interface dynamics, free volume estimation and the effect of intermolecular interactions and hydrogen bonding are discussed. The effect of these results on electrical property such as dielectric strength (DES) at room temperature was studied.

1.1. Multi-core interface model

The interaction of nanoparticles with the surrounding polymer matrix by means of three layers is described by Multi-core model [1] as shown in **Figure 1**. It consists of (i) Primary layer also referred as bonded layer, (ii) Secondary layer as referred as called bound layer, (iii) tertiary layer also referred as called loose layer, and the next fourth layer which overlaps all the above three layers called electric double layer. Primary layer represents a type of transition layer which is firmly attached to the carbonless nanofiller or inorganic nanofiller and carbon based organic base matrix polymer by compatibilizer or hardner. Secondary or bound layer addressed as interfacial layer or region consists of a region or area of layer of polymer chains

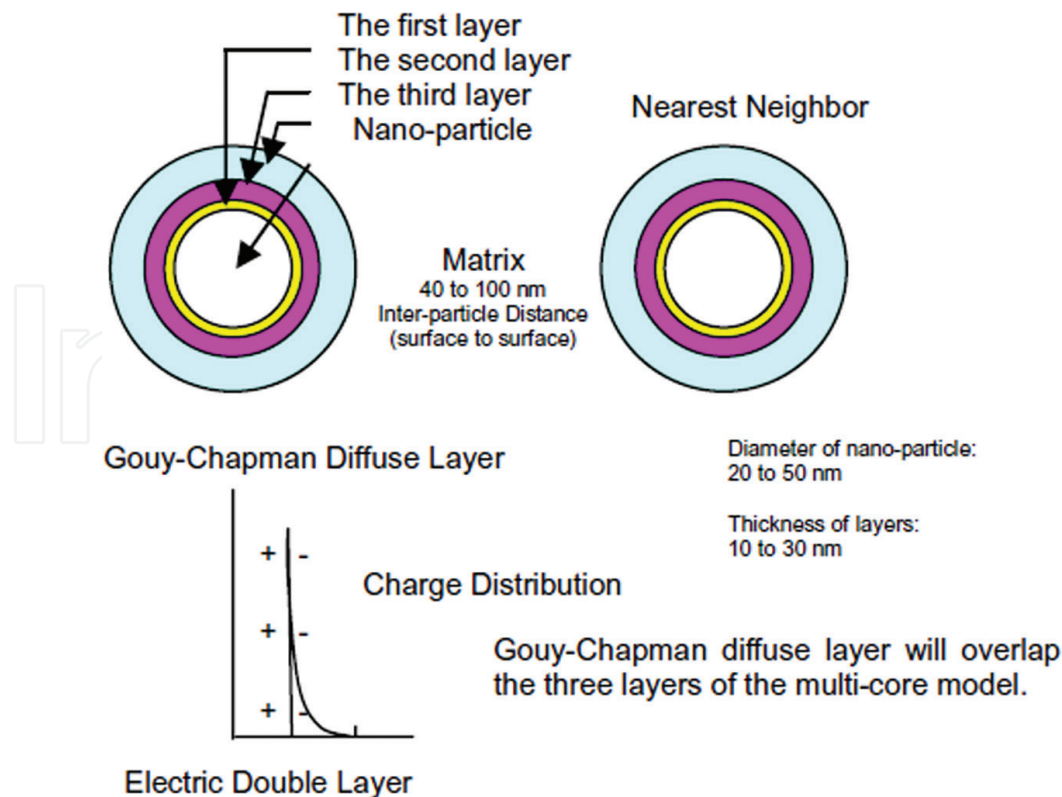


Figure 1. Multi-core model for nano-particle-polymer interfaces (source: Toshikatsu Tanaka and co-authors [1]).

strongly bound to the primary layer and outer surface of nanofiller. Thickness of the layer varies between 2 and 9 nm. This value mainly relies on the strength of interaction between organic polymer and nanofiller or nanoparticles. Stronger the interaction, the larger will be the bound polymer fraction. This may correspond to a stoichiometrically cross-linked layer.

The loose layer is a region which is loosely coupled, at the same time interacts with the bound region. It is generally considered that the loose layer has different chain conformation, chain mobility, and even free volume in the polymer matrix. It may also consist of a less stoichiometrically cross-linked layer. In addition, interfacial structures obtained from chemistry, Columbic interaction is superimposed, when dielectric and electrical insulation properties are investigated [1]. The nanoparticle may be charged either positively or negatively. When a polymer has mobile charge carriers, they are distributed in the interface in such a manner that the charge carriers with the opposite polarity are diffused outward from the contact surface to the Debye shielding length that corresponds to the Gouy-Chapman diffuse layer in which charge decays exponentially with distance, in accordance with Born approximation [1]. Debye shielding length is calculated approximately as 30 nm [1].

The thickness of these layers may varies from 1 nm to, several tens nm for the bonded, bound and loose layers respectively. It is not clearly known whether the thickness of the loose layer is the same as that of the Gouy-Chapman diffuse layer. It may appear that the latter might extend over the former. Therefore, far-field effect must be involved in mesoscopic interactions in the loose layer or the diffuse Gouy-Chapman layer, and it is expected to cause some combined effect among neighboring nanoparticles. Macroscopic phenomena and parameters are different from polymer to polymer and polymer with filler particles due to the relative differences in its thickness and interaction strengths in the multi-core model with the far-field effect.

2. Experimental studies

2.1. Matrix and fillers

The Bisphenol A diglycidyl ether based Epon 828 epoxy resin (DGEBA) with epoxy equivalent weight (EEW) of 188 g/mol and curing agent such as Epikure W which is chemically called diethyl toluene diamine (DETDA) with an amine hydrogen equivalent weight of 45 g/mol were used in the present work. These materials were supplied by M/s. Miller-Stephenson Chemical Company, USA. The nanoclay used in the present work is called as Nanomer 1.30E supplied by M/s. Nanocor, USA. This nanoclay was surface treated with surface functionalizer namely octadecylamine mainly used for uniform dispersion of nanoparticle epoxy resin polymer. This surface functionalized nanoclay is called organically modified montmorillonite clay represented as oMMT.

2.2. Fabrication process of nanocomposite

One of major challenges in the processing of nanocomposites is the non-uniform mixing of curing agent. Non uniform mixing of nanofiller, resin, and the curing agent or hardner may

also results in an improper curing. The schematic diagrams of processing method and curing cycle are shown in **Figures 2** and **3** respectively.

Steps for processing of epoxy-oMMT nanocomposites:

Step1: The viscosity of Epon 828 resin was high at room temperature; and therefore, it was difficult to mix. In order to reduce the viscosity before mixing, the resin was preheated in an oven at 60°C for about 2 hours. The filler was dried in an oven at 100°C for 24 hours.

Step2: After reducing the viscosity, a known weight of epoxy resin was taken.

Step3: The required weight of the oMMT and curing agent were added to the epoxy resin.

Step4: Epoxy resin, oMMT, and curing agent mixture were mixed using IKA high shear mixer (T-T18 ULTRA TURRAX Basic) at a speed of 24,000 RPM for 45 minutes.

Step5: After mixing, degassing was carried out in a vacuum oven for 45 minutes.

Step6: The, the mixture was then transferred into aluminum molds and degassed again for 30 minutes. After degassing, the aluminum molds were placed in an oven, and the materials were cured based on the time–temperature curing cycle shown in **Figure 3**. The dimensions of cured sheet of epoxy-oMMT nanocomposites used for the investigation had an area of 200 mm × 200 mm and thickness of 3 mm.

2.3. Measurements

2.3.1. Interface dynamics

(i) Fourier transform infrared spectroscopy (FTIR)

Nanoparticles chemistry and chemical bonding type that existed between polymer and nanoparticles of cured polymer nanocomposites in the present study were characterized through FTIR measurements.

(ii) Differential scanning calorimeter (DSC)

The nanocomposites melting and glass transition temperature were calculated using DSC model 821 of Thermal Analysis instruments.

(iii) Positron annihilation lifetime spectrometer (PALS)

The PAL spectra of the nanocomposites have been traced by means of positron lifetime spectrometer with time resolution of 220 picoseconds.

(iv) Dielectric strength (DES)

The electrical breakdown measurements were carried out using HV AC Test Set of M/s. W.S. Test Systems Pvt. Ltd., Bangalore.

2.4. Experimental techniques

The bonding of nanoparticles with the base polymer and the chemical nature of cured polymer material were characterized through FTIR measurements using Perkin Elmer make, model spectrum-GX FT-IR as per ASTM D 7214-07a.

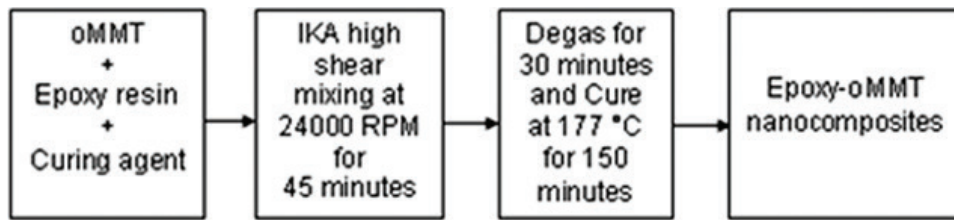


Figure 2. Processing of epoxy-oMMT nanocomposites.

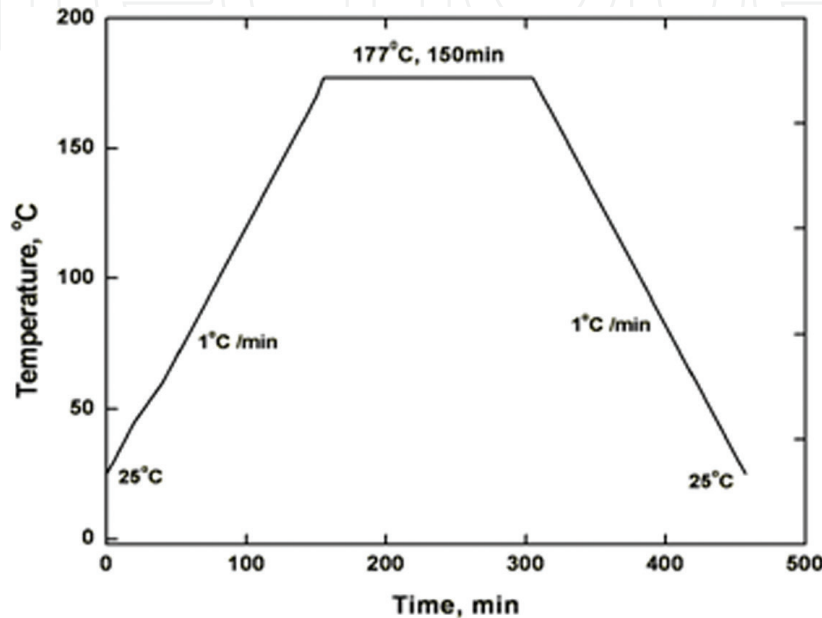


Figure 3. Curing cycle for epoxy-oMMT nanocomposites.

The DSC Model 821 of Thermal Analysis instrument was operated in nitrogen atmosphere to determine the glass transition temperature as per ASTM D-3428-99. The PALS in pure epoxy resin and oMMT filled nanocomposites were recorded using the positron lifetime spectrometer with a time resolution of 220 ps [2]. The PAL spectra obtained were analyzed by employing the computer software PATFIT [3]. This software decomposes a PAL spectrum into three discs. The lifetime component τ_2 with intensities I_2 is due to trapping of positrons at the defects. The longest life components τ_3 with intensity I_3 is due to pick-off annihilation of the o-Ps from the free volume sites present mainly in the amorphous regions of the polymer matrix. Nakanishi and co-authors [4] proposed equation 1, which is referred to the previous research works of Tao [5] and Eldrup [6] utilized to compute radius(R) of the free volume cell from the noted values of τ_3 (o-Ps lifetime).

$$\frac{1}{\lambda_3} = \tau_3 = 0.5 \left[1 - \frac{R}{R + \Delta R} + \frac{1}{2\pi} \sin\left(\frac{2\pi R}{R + \Delta R}\right) \right]^{-1} \quad (1)$$

Here, ΔR is the fitting parameter and has been found to be 1.656 Angstrom (\AA) for solid molecular media [7]. The free volume size is evaluated as $V_f = (4/3) \pi R^3$.

The fractional free volume or the free volume content (F_v) is calculated as $F_v = CV_f I_3$, where, $C = 0.00018 \text{ nm}^3$ [7, 8].

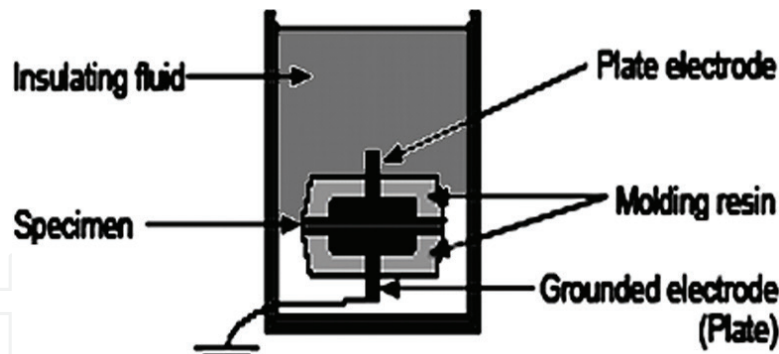


Figure 4. Insulation breakdown test method by needle-plate electrode geometry.

The electrical breakdown voltage was measured at 300.15 K as per ASTM D-149 as shown in Figure 4.

Electrical breakdown voltage of nanocomposite insulator was documented. The dielectric breakdown strength (DES) was computed using,

$$E = \frac{V}{t} \text{ KV/mm} \quad (2)$$

here, V = Electrical breakdown voltage in kilo volts,

t = thickness of nanocomposite sample in m.

3. Results and discussion

3.1. FTIR spectrum of pure epoxy

FTIR spectrum of pure epoxy is shown in Figure 5. In the below figure, peaks corresponding to the presence of functional groups in the epoxy system and are listed in Table 1.

The epoxy resin contains several polar groups which can interact with the surface $-OH$ groups on the nanoparticles through hydrogen bonding rather easily, similar to the bonding between the $-OH$ groups and H_2O molecules in the atmosphere. An uncured DGEBA epoxy resin is polar in nature and contains two epoxide groups at both ends.

3.2. Hydrogen bond formation in polymer and nanoparticle

In the fabrication process of the epoxy nanocomposites, the filler (α MMT) is first mixed with DGEBA based epoxy subsequently the addition of hardner (DETDA) into the epoxy and α MMT nanoparticle mix to initiate the curing process. When DETDA hardner is added to the epoxy and α MMT particle mix, the epoxide group open up and form hydrogen bond with the free $-OH$ groups on the nanoparticle surface in addition to reacting with amine groups of the hardner as shown in Figure 6.

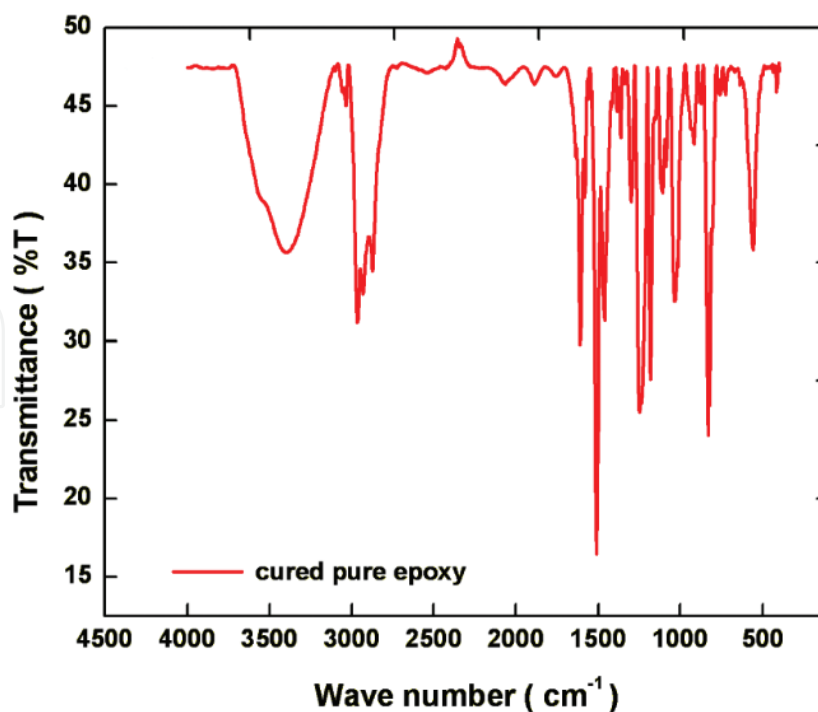


Figure 5. FTIR spectra of pure epoxy.

Wave number (cm ⁻¹)	Functional groups
~3399	OH groups
~3036	Corresponds to the C-H stretch in aromatics
~2965	Corresponds to asymmetrical C-H stretch of -CH ₃ group
~2932	Corresponds to asymmetrical C-H stretch of -CH ₂ group
~1608	} Corresponds to C-C stretching vibration in aromatic
~1582	
~1508	
~1458	
~1298	Corresponds to asymmetrical -CH ₂ deformation
~1247	Corresponds to asymmetrical aromatic C-O stretch
~1181	Corresponds to asymmetrical aliphatic C-O stretch
~1085	Corresponds to symmetrical aromatic C-O stretch
~916	Corresponds to epoxide ring vibrations
~874	Corresponds to -CH out of plane deformation in aromatic
~560	

Table 1. FTIR peaks corresponding to functional groups in pure epoxy.

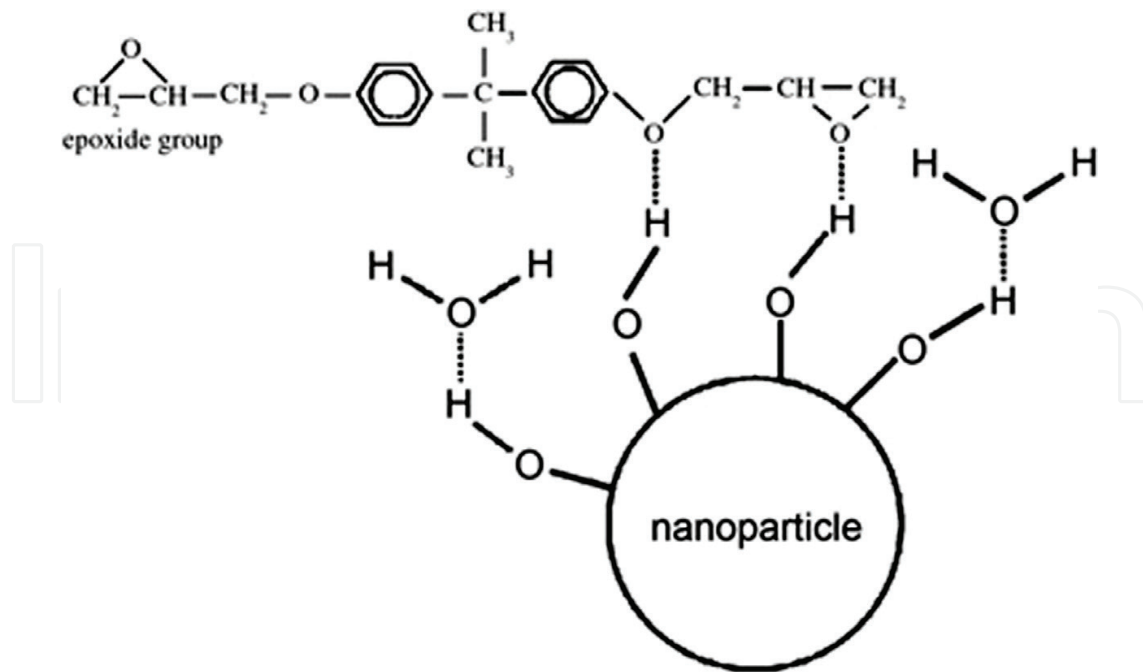


Figure 6. Hydrogen bonding between $-OH$ groups on the nanoparticle surface and epoxide groups in epoxy during curing.

Since, there is abundantly free $-OH$ groups are available on the nanoparticle surface, the hydrogen bonded epoxy segments will be more in number nearer to the nanoparticle surface. Development of the hydrogen bonds at interface region of epoxy and nanofiller is due to strongly bonded first nanolayer and tightly bounded second nanolayer of nanofiller and epoxy segments. This is in accordance with multi core model [1].

3.3. FTIR spectra of epoxy-oMMT nanocomposites

The FTIR spectrum of epoxy-oMMT nanocomposites is shown in **Figure 7**. The FTIR spectra clearly show that the incorporation of the filler particles leads to formation of hydrogen bonds. The interaction mechanism between nanofiller particles and base epoxy is the formation of hydrogen bond with free surface hydroxyl ($-OH$) groups present on the nanofiller particle outer surface and the base polymer epoxy. One of the major understand with the hydrogen bonding is that they will not exhibit any such new peaks in FTIR spectra.

The band at 3444 cm^{-1} correspond to the stretching vibration of hydroxyl ($-OH$) groups attached to the nanoparticle surface. The $-OH$ groups may be attached on the surface of nanofiller particles as free $-OH$ groups, or as $-OH$ groups attached to absorbed H_2O molecules. When the band appears at 1638 cm^{-1} , this corresponds to the bending vibrations of absorbed H_2O molecules. The presence of the FTIR peaks at the above mentioned wavelengths reports the presence of hydroxyl ($-OH$) groups on the nanofiller surfaces. This result obtained through FTIR measurements in the present work have been well supported by several researchers [9–11].

The reason for the presence of H_2O molecules on the surfaces of oMMT particles are affinity of H_2O molecules present in atmosphere to get bonded to the surface OH groups on the oMMT particles through hydrogen bonding. H_2O is a polar particle or molecule and O_2 attached to the

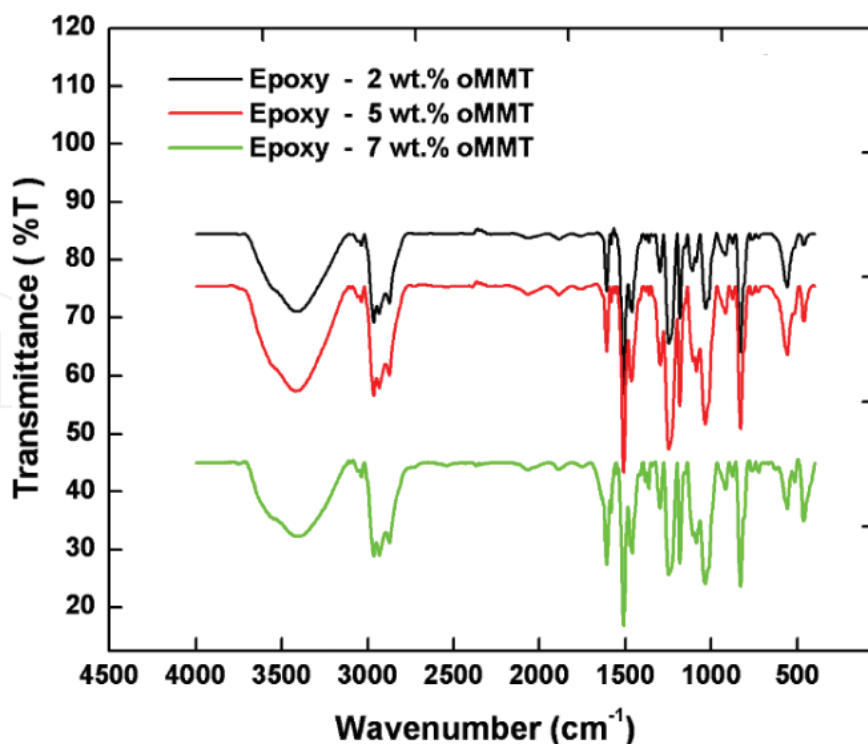


Figure 7. FTIR spectra of epoxy-oMMT nanocomposites.

H₂O molecule is highly prone to hydrogen bonding with free hydroxyl groups present on the OMMT particle surfaces as shown in Figure 7. Hence, there will be a tendency for all the –OH groups present in the oMMT nanoparticle surface to form hydrogen bonds with H₂O molecules in the atmosphere which may result in the formation of a H₂O layer on the surface of the OMMT particles. When such oMMT particles having high concentration of surface H₂O molecules are introduced into epoxy, the H₂O molecules will tend to influence the thermal and electrical properties of the epoxy based nanocomposites [12]. Although, it is very difficult to fully remove the formation of hydrogen bonds between the surface –OH groups and H₂O molecules, the experiments carried out have to be controlled to reduce the formation of a H₂O layer on the surface of the nanoparticles. One of the important processes which has been adopted in the present work is to adequately dry the OMMT particles before they are adding into the polymer matrix.

With reference to the Figure 7, the width of the peak, intensity of the peak and –OH infrared peak positions are influenced by the existence of hydrogen bonding in the oMMT nanofiller and epoxy matrix. However, it is well known that for –OH bands i)width of peak, and ii) intensity of the peak increases as the incorporation of oMMT particles in epoxy resin increases which is considered as sign of adding up of hydrogen bonds in the nanofilled filled composites.

3.4. Analysis of the glass transition temperature (T_g)

The variations in T_g of the epoxy nanocomposites are shown in the Figure 8. It is noted from the figure, about the T_g value of pure epoxy resin as 442.15 K. It is also noted from the plot that inclusion of 2 wt.% oMMT, into epoxy results in increased value of T_g by 278.15 K as compared to pure epoxy.

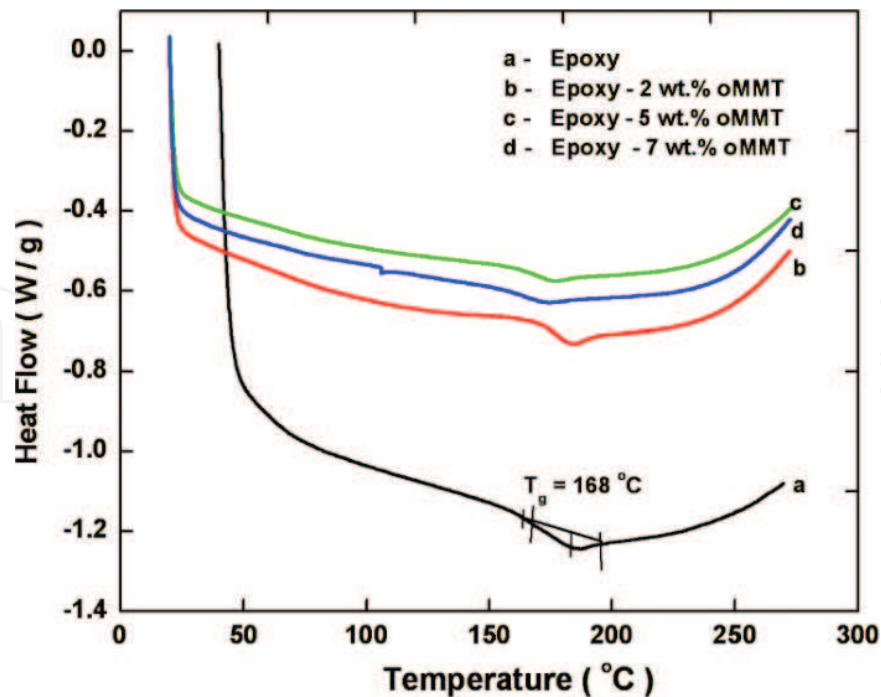


Figure 8. DSC plots of epoxy-oMMT nanocomposite.

With the incorporation of 5 and 7 wt.% of nanofiller T_g value decreases. The interfacial interaction of polymer nanocomposites and following in the formation of nanolayer in nanocomposites are concluded in the literature followed. The formation of nanolayer and its influence on T_g is also been reported. The higher value of in T_g [13, 14] when nanofiller has been added is reported in some cases whereas, some others report a smaller value of T_g due to addition of nanoparticles into base polymer matrix [15], but the majority of the proposed theories are uncertain till today. The influence of oMMT nanofiller on the curing reaction and glass transition values of the epoxy nanocomposites is investigated by DSC in order to understand the molecular mobility in the nanocomposites. When the nanofiller concentration is increased from 5 to 7 wt.%, the values of T_g in epoxy nanocomposites is observed to decrease. Similar to the results obtained in the case of epoxy filled with oMMT nanofillers in the present work, reduced value of T_g have been reported for nanoalumina filled PMMA composites [16]. The increase in T_g with the addition of 2 wt.% of nanofiller is due to few cross links that developed between the polymer matrix and nanoparticle. However, the reduction in T_g values at 5 and 7 wt.% of nanofiller loading may be due to the many reasons such as changes in molecular weight, tacticity, cross-linking density and the presence of residues from incomplete reactions. The other possible reasons could be the size of the nanofillers which are certainly larger than the free volume hole sizes (discussed in Section 2.4) in the matrix and therefore the possible slide between the chains can result in increased free volume.

Sun and co-authors [17] reported that the T_g depression is associated with the improved polymer chain dynamics because of the additional free volume at the resin-nanofiller interface. Becker and co-authors [18] remarked, interfacial interactions between polymer chains and positively or negatively charged nanofiller surface leads to the development of a polymer

nanolayer near to the nanoparticle surface and this interfacial polymer nanolayer influences the T_g . These nanoparticle and polymer interactions can be developed as attractive interaction, repulsive interaction or the interaction is neutral and as such T_g can increase, decrease, or remain constant. In the present work, the results obtained for 2 wt.% nanocomposites, may be due to few cross links but for higher loading, the nanoparticle-polymer interaction found to be repulsive and therefore T_g decreases and the present results are in good concord with the published literatures [16, 17]. The cross linking density reduces due to etherification mechanism and curing agent preferably tends to attach to the surfaces and very thin layer of hardner or curing agent will surround the nanofiller particles. This thin layer of curing agent will keep the curing agent around the nanofiller particles from reacting with the epoxy resin. This 'curing agent concentration' mechanism will also cause decrease in cross-link density and thereby T_g decreases at 5 and 7 wt.% of filler loading.

3.5. Characterization of free volume content

The variation of free volume content or fractional free volume (F_v), free volume size (V_f) and longest life time (τ_3) with 2, 5 and 7 wt.% of oMMT in epoxy nanocomposites is shown in Figure 9.

The addition of oMMT content in epoxy resin (2-5 wt.%), causes τ_3 to increase from 1.72 to 1.75 ns, that is size or area of the free volume and free volume content increases from 7.17 to 7.45 nm³ and 2.28 to 2.45% respectively. However, at 7 wt.% oMMT loading, both τ_3 , V_f and F_v decrease slightly to 1.74 ns, 7.34 nm³ and 2.14% respectively. The modifications has taken place may be because of addition of nanofiller in polymer matrix creates additional free volume. It further suggests that, the filler will not occupy the pre-existing free volume cavities due to

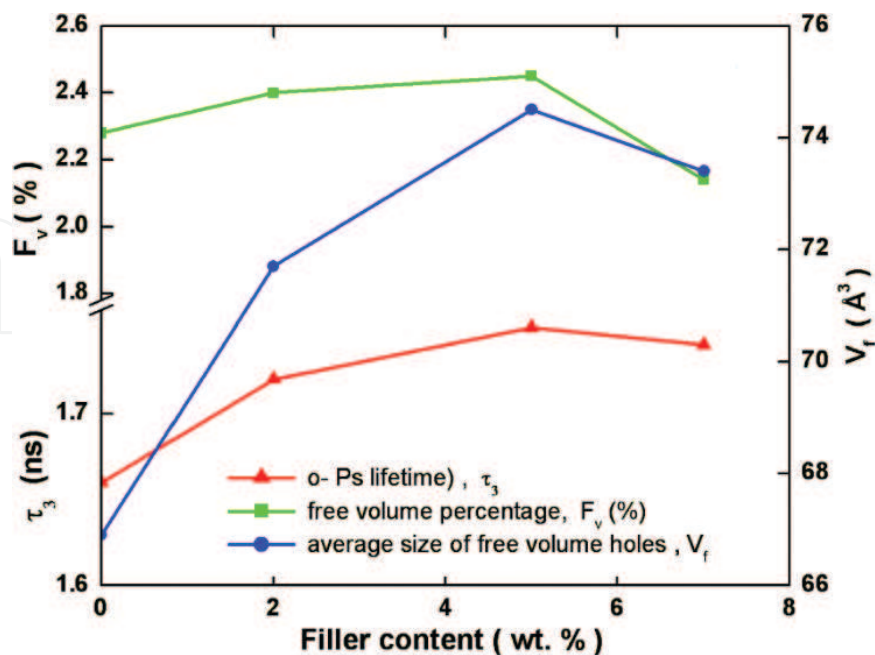


Figure 9. Plot of τ_3 , V_f and F_v versus filler content (wt.%) of epoxy-oMMT nanocomposites.

bigger size of the fillers but creates additional free volume probably at the interface. The results also indicate that the layers of oMMT will result in favorable interaction with the epoxy resins and thereby the segmental motion is hindered. Hence, the significant increase in F_v percentage is not observed. The decrease in T_g is justified by the decrease in free volume content.

3.6. Effect of interface on dielectric strength (DES)

The dielectric strength (DES) is the vital properties of dielectric insulators. With reference to the discussion in Section 2.4, the properties of epoxy nanocomposites are mainly explained by interfacial interaction of polymer and of nanofillers. This interfacial area is responsible for the interaction of the electric field between the base epoxy and nanofiller. The DES of the nanocomposites depends largely on nanofiller content and even a very less quantity of nanofiller can cause improvement. When nanofiller particles are incorporated into the epoxy matrix, there is a change in morphology of the epoxy due to the interfacial interaction of epoxy with the oMMT nanofiller.

The **Figure 10** shows the variation of Dielectric strength, free volume with respect to nanofiller loading. With the addition of 2 wt.% oMMT into epoxy matrix, it is observed that T_g increases due to increase in cross linking density and hence less free charge carriers are available leading to slightly higher value of DES than that of pure epoxy. A further increase in the filler loading up to 5 wt.% of oMMT shows an increase in the DES above that of 2 wt.% of nanocomposite. In this case the T_g decreases, and hence more free charge carriers are available.

Here the effect of third interface layer also called loose layer comes into scenario. The loose polymer layers contain more traps or free volumes as discussed in Section 2.4. These charge carriers are easily and more frequently trapped in trap sites rather than in the base epoxy.

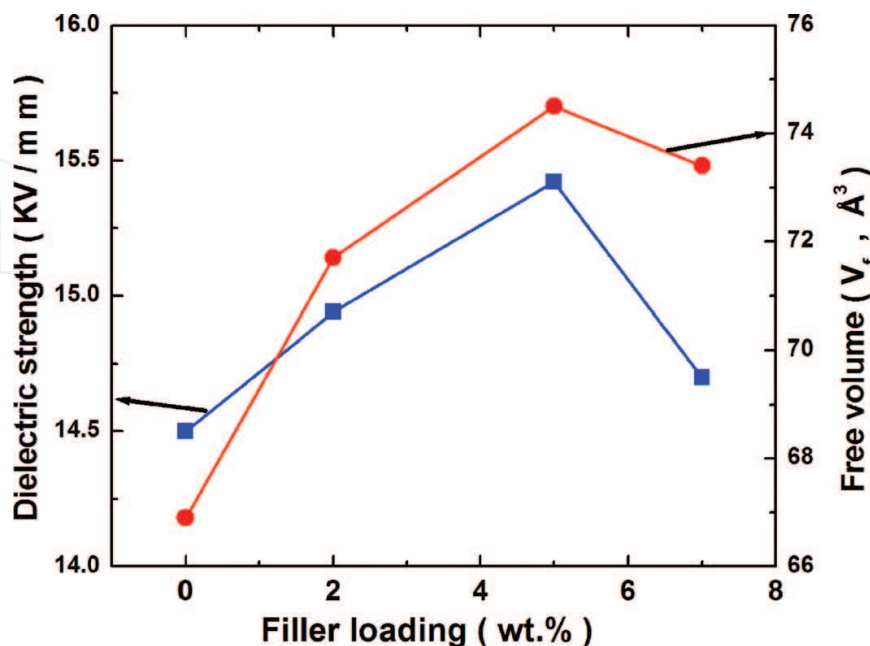


Figure 10. Plot of DES, Fv with respect to filler content (wt.%) of epoxy-oMMT nanocomposites.

Because of this cause, charge carriers are accelerated over shorter distances and have reduced mobility and kinetic energy. This process is considered as a scattering mechanism. The energy of charge carriers is distributed more evenly in the polymer and thus causes less damage in the material and prolongs the lifetime and service of the epoxy. This scattering process decreases the electric field at the electrodes and increase the voltage required for charge injection.

These fillers can also act as barriers for the penetration of the charge carriers throughout the depth of the sample. In case of 5 wt.% oMMT, the number of nanoparticles is much more and the inter particle distance is also less than 100 nm. This inter particle distance was justified by morphological studies. Hence, there is a possibility of overlapping of the loose polymer regions in the nanocomposites leading to the reduction in the loose polymer regions. This polymer layer along with a large number of fillers can also obstruct the discharge path, thereby dielectric strength increases. The increase in dielectric strength values is well supported by reduction in dielectric constant values and increase in free volume content at 2 and 5 wt.% oMMT filled epoxy-oMMT.

When oMMT filler content is increased to 7 wt.%, there is a reduction in the dielectric strength occurs due to the overlapping of the tightly bound polymer regions over the interface, since the inter particle distances are comparable to the filler diameter. The interphase region close to the nanoparticle is found to be conductive [11]. As the conductive interphase regions tends to overlap, the pure polymer region is reduced, leading to an easier conducting path for the charge transfer and thereby a reduction in the dielectric strength is observed. With the addition of 7 wt.% of oMMT into epoxy matrix, reduction in T_g has taken place, and hence more free charge carriers are available, but reduction in free volume leads to an easier conduction path for the charge carriers and thereby DES decreases. The reduction in DES at 7 wt.% of oMMT filled nanocomposite is well supported by increase in dielectric constant values and free volume content of the nanocomposites. This has been well supported by many authors. Many researchers [19] have reported that layered silicate nanofillers modify the trapping property of both isotactic and syndiotactic PP. Roy and co-authors [20] have reported that deep trap sites have been identified in SiO_2 -XLPE nanocomposites through thermally stimulated current (TSC) measurements.

4. Conclusions

The following remarks are drawn:

- i. Existence of hydrogen bond between nanofillers and epoxy polymer chains in the nanocomposites has been established through FTIR, T_g , and free volume measurements.
- ii. The interaction takes place between epoxy and nanoparticle in small region around the oMMT nanofiller surface called as "interface region" and the nature of interaction is found to be depends on the chemical bond of the oMMT and epoxy.
- iii. The interaction between the oMMT and epoxy chains has a direct effect on dielectric strength characteristics of the nanocomposite and accordingly, a three core interface model has been used to elucidate the characteristics of the interface region.

- iv. The characteristics of the interface region of the nanocomposite depend on the number of oMMT nanofiller particles included in the epoxy resin and how the oMMT nanoparticle loading affects the dielectric strength.

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