Investigation on the impact of nano MgO addition on dielectric properties and space charge behavior of epoxy resin AlN nanocomposites

Guvvala Nagaraju and Ramanujam Sarathi

Department of Electrical Engineering, Indian Institute of Technology Madras, Chennai 600036, India

* Author to whom any correspondence should be addressed.

E-mail: rsarathi@iitm.ac.in

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Abstract

Epoxy nano composites were fabricated with the incorporation of nano AlN and MgO fillers to investigate their dielectric and space charge properties. Epoxy AlN nanocomposites with the addition of nano MgO have shown lower permittivity with reduced tanδ, good space charge behaviour, during poling period. Higher wt% of AlN in epoxy resin exhibited higher permittivity. The nanocomposites with AlN nanoparticles have revealed higher space charge accumulation than on addition of MgO nano filler. Also, the addition of nanofillers to the base polymer has led to enhanced space charge decay performance compared to the base matrix.

1. Introduction

Epoxy resin (ER) is widely used as insulating material in power apparatus because of its excellent electrical, thermal, mechanical properties with good chemical stability [1]. In recent times, epoxy nanocomposites formed by addition of inorganic oxide nanoparticles as filler to the base epoxy have shown novel properties and considered as futuristic material with high mechanical strength, enhanced thermal conductivity with increased dielectric strength. With the increased rating of the power devices, it has become essential to have insulation structure with good dielectric properties with high thermal conductivity. In addition, under DC voltages, it has become essential to have space charge free insulation structure [2]. The world over researchers have indicated that use of magnesium oxide (MgO) nano filler can effectively relieve the space charge formation in the bulk insulation structure [3–6]. Aluminium nitride (AlN) is one of the most widely used nano fillers for enhancing the thermal properties of the base polymer. It is also reported that, adding one or more filler of micro/nano and nano form to the base polymer can provide better performance [7–9]. The literature on the space charge performance of nano mixtures composites is limited.

With the above review of literature, a methodical experimental study was carried out, to understand the impact of addition of MgO nanoparticles with AlN nano particles to the epoxy resin on its variation to (a) Permittivity and tanδ of the epoxy nanocomposites and (b) space charge analysis adopting Pulsed electro acoustic technique.

2. Experimental setup

2.1. Samples preparation

Epoxy nanocomposites were prepared with MgO and AlN as nano fillers. Table 1 shows composition of nano filler in each type of nanocomposites. The epoxy resin (CY 205–Huntsman, India Pvt. Ltd.) was used as the base polymer and Tri-ethylene tetra-amine (HY 951–Huntsman, India Pvt. Ltd.) was used to cure the epoxy resin. For the preparation of first and second set of specimens, the nano fillers were dried for 5 h in air circulated oven, by maintaining at 150 °C to remove the traces of moisture present in the nano powder. The required quantity of
nano powder was mixed with ethyl-alcohol (solvent) and kept in ultrasonic bath for 1 h and then added to the epoxy resin. Added mixture was mixed by using high speed shear mixer for 60 min and then through ultra-high frequency sonication process (20 kHz) for 60 min, followed which the mixing process was continued for 8 h by high speed shear mixing and kept for ultra- high frequency sonication for 60 min. At the end, the hardener was mixed and degassed and then final mixture is casted in to a steel mould. For third set of samples, required quantity of MgO and AlN nano filler mixed with the solvent and was added to the epoxy resin. The procedure adopted for preparing epoxy resin with single nano filler was followed with the ternary process, to prepare epoxy nanocomposite sheets. Figure 1 shows steps adopted for preparation of nanocomposites.

2.2. Space charge measurement
Pulsed electro acoustic technique was adopted for space charge measurement as shown in figure 2. The PEA experimental setup is equipped with a DC high-voltage (0–30 kV) source, a 0–500 V range pulse generator source with operating frequency as 150 Hz and generates pulses with width of 10 ns. The high-voltage and ground were made up of brass and aluminium materials respectively. A sensor made up of Polyvinylidene fluoride (PVDF) material is used for acoustic signal detection and an amplifier was used to amplify the detected pulse acoustic signal. A digital storage oscilloscope (Tektronix, DPO-5034B) with PEA signal acquisition

![Figure 1. Steps for epoxy Nanocomposites preparation.](image1)

![Figure 2. Experimental setup for space charge measurement.](image2)

| Sample | MgO | AlN | Sample | MgO | AlN |
|--------|-----|-----|--------|-----|-----|
| E      | 0   | 0   | A3     | 0   | 3 wt% |
| M1     | 1 wt% | 0   | A5     | 0   | 5 wt% |
| M3     | 3 wt% | 0   | M2A1   | 2 wt% | 1 wt% |
| M5     | 5 wt% | 0   | M1A2   | 1 wt% | 2 wt% |
| A1     | 0   | 1 wt% | —     | —   | —   |
The software was connected to the PEA cell for signal recording. The poling (voltage-on) and depoling (voltage-off) space charge measurements were performed in the present study, each for 1 h.

2.3. Dielectric response measurement

Dielectric relaxation spectroscopy (DRS) analysis of the specimens was carried out to measure the complex permittivity of the material. The dielectric measurements were carried out by using broadband dielectric-impedance spectroscopy analyzer in the frequency range of 0.1 Hz to 1 MHz, at room temperature. 1.5 mm thick, 30 mm diameter samples were used for the study.

3. Results and discussion

3.1. Dielectric properties

Figure 3 shows the variation in permittivity ($\varepsilon'$) of the epoxy nanocomposites at different frequency. It is observed that M1 specimen showed increased permittivity and M3 specimen showed lower permittivity compared to neat epoxy resin. Specimen M5 shows higher permittivity in the lower frequency range as shown in figure 3(a). An increase in permittivity for 1 wt% MgO filled epoxy nanocomposite is observed compared to neat epoxy, which could be due to the influence of filler permittivity. With further increase in the nanofiller concentration, an increase in interfacial area between filler and matrix occurs reducing the mobility of the polymer chain bonding to nano particle causing reduction in the relative permittivity of the material [10–12]. Du et al also observed similar variation in permittivity of epoxy nanocomposites filled with TiO$_2$ nanoparticles [13]. Variation in relative permittivity of epoxy AlN composites is shown in figure 3(b). It is observed that its relative permittivity is high compared to the neat epoxy specimen. In polymer nanocomposites, the filler size have great influence on the effective permittivity of the material [14]. In the present study, AlN nanoparticles have an average particle size of $<100$ nm, which is 4 to 5 times higher compared to MgO nanoparticles (average particle size, 20 nm), which could be the cause for increased relative permittivity. The variation in relative permittivity of MgO co-added AlN epoxy nanocomposites is shown in figure 3(c). It is observed that M2A1 specimen has least permittivity among all epoxy nanocomposite specimens. Whereas, M1A2 has showed similar permittivity variations compared to the neat epoxy specimen E, except at lower frequencies. The dielectric response of the ternary composites are surprising and further investigations have to be done to understand the synergic effect of two nanofiller with the epoxy matrix.

Under low frequencies the dielectric loss of the MgO filled epoxy nanocomposites is higher than the neat epoxy specimen E, whereas, at high frequencies (>1 kHz) the specimen M3 has less dielectric loss compared to all the specimens as shown in figure 4(a). MgO filled epoxy nanocomposites showed broad $\beta$ relaxation peak at higher frequencies, because of the restricted side chain movement of epoxy network which were responsible for $\beta$ relaxation [14]. Hence, with MgO filled epoxy nanocomposites a slight shift in the $\beta$ relaxation peak was observed. It can be seen that all AlN epoxy nanocomposites showed increase in dielectric loss under lower frequencies and at higher frequencies (>10 kHz) the dielectric loss was almost near equal to that of the neat epoxy E as shown in figure 4(b). The interfacial area around MgO nanoparticle will be high in comparison to that AlN filler. This could be the reason for the epoxy nanocomposites filled with AlN nanoparticles having less interfacial influence on the chain mobility exhibits higher dielectric loss under low frequencies. From figure 4(c) it can be seen that compared to neat epoxy specimen E, ternary composite M2A1 has higher dielectric loss at lower frequencies and reduced dielectric loss at higher frequencies. With increase in the wt% of AlN filler in
hybrid epoxy nanocomposites, specimen M1A2 has showed more dielectric loss compared to unfilled specimen E. Overall, M2A1 epoxy nanocomposite has showed good dielectric response.

3.2. Space charge characteristics

3.2.1. Space charge accumulation

The typical space charge profiles of epoxy nanocomposite specimens under an applied electric field of 10 kV mm⁻¹ are shown in figure 5. Hetero charge formation was observed near electrodes for neat epoxy specimen E, as shown in figure 5(a). Figure 5(b) shows accumulation of little amount of hetero charge near the both electrodes and considerable amount of positive charge within the bulk of the specimen M5. Figure 5(c) shows both positive and negative charge accumulation within the bulk of the specimen M5. It is noticed that very less space charge for the specimen M2A1 as shown in figure 5(d).

It can be realized that from space charge accumulation studies, MgO co-filled AlN epoxy nanocomposites with higher MgO filler content has improved insulation performance compared with AlN/MgO filled epoxy nanocomposites. The absolute amount of accumulated bulk charge density in the specimen was calculated based on the following equation [15].

![Figure 4. Variation in dielectric loss of (a) MgO filled (b) AlN filled and (c) MgO co-addition AlN epoxy nanocomposites.](image)

![Figure 5. The dynamics of space charge distribution in sample (a) E (b) M5 (c) A3 and (d) M2A1 epoxy nanocomposites.](image)
\[ q(E, t) = \frac{1}{L} \int_{x_0}^{x_1} |q_s(x, t)| \, dx \] (1)

Where, \( L \) is the thickness of the specimen, \( x_0 \) and \( x_1 \) represents the electrode positions and \( q(x, t) \) is the measured space charge density at a distance \( x \) from the cathode during time \( t \). Figure 6 shows the average amount of accumulated charge density during poling period. It was observed that, irrespective of type and wt% of nanofiller used, the accumulation of space charge within the bulk of the insulation and reaches saturation. After 1 h of poling, the average amount of accumulated space charge density is high with M1 sample and least with M2A1 specimen.

Figure 6. Accumulation of space charge during poling with epoxy nanocomposites.

Figure 7. Typical variation in space charge decay during depoling period for epoxy nanocomposites (a) E (b) M5 (c) A3 and (d) M2A1.
3.2.2. Space charge decay
The typical space charge decay profiles of the different specimens after volt off (VO) is shown in figure 7. The dynamics of space charge decay is slow in base epoxy as shown in figure 7(a). With increase in wt% of MgO content in epoxy nanocomposites, a reduction in the hetero charge formation near the cathode was observed (figure 7(b)). In epoxy AlN nanocomposites homocharge was observed immediately after the voltage off and negative charge within the bulk, as shown in figure 7(c). After voltage off the total charge from M2A1 specimen was disappeared in less than 100 s as shown in figure 7(d).

The space charge density decay of all the specimens on switching off the voltage after 1 h of poling are shown in figure 8. It can be seen that decay of space charge was fast within nanocomposites compared to neat epoxy specimen E except for specimen M1. The space charge decay rate of the specimens was nonlinear with time. It can be seen that immediate after switching off the voltage the decay rate was high and with time the charge decay rate decrease. Addition of nanofillers to the base polymer showed enhanced space charge decay compared to the base matrix.

4. Conclusion
The important conclusions arrived at based on the present study are the following.

- Epoxy ALN nanocomposites on addition of nano MgO showed lower permittivity with reduced tanδ, good space charge behaviour, during poling period. Higher wt% of AlN in epoxy resin showed higher permittivity.

- AlN nanofiller added nanocomposite samples shows higher space charge accumulation than on addition of MgO nano filler. Also, addition of nanofillers to the base polymer showed enhanced space charge decay performance compared to the base matrix.

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Data availability statement
The data that support the findings of this study are available upon reasonable request from the authors.
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