Effect of Space Charge Density and High Voltage Breakdown of Surface Modified Alumina Reinforced Epoxy Composites

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The incorporation of 90 nm alumina particles into an epoxy matrix to form a composite microstructure is described in present study. It is shown that the use of ultrafine particles results in a substantial change in the behavior of the composite, which can be traced to the mitigation of internal charges when a comparison is made with conventional Al2O3 fillers. A variety of diagnostic techniques have been used to augment pulsed electro-acoustic space charge measurement to provide a basis for understanding the underlying physics of the phenomenon. It would appear that, when the size of the inclusions becomes small enough, they act cooperatively with the host structure and cease to exhibit interfacial properties. It is postulated that the Al2O3 particles are surrounded by high charge concentrations. Since Al2O3 particles have very high specific areas, these regions allow limited charge percolation through Al2O3 filled dielectrics. The practical consequences of this have also been explored in terms of the electric strength exhibited. It would appear that there was a window in which real advantages accumulated from the nano-formulated material. An optimum filler loading of about 0.5 wt.% was indicated.

Keywords: Epoxy composite, Dielectric properties, Electrical insulation breakdown strength

1. INTRODUCTION

Insulating polymers are widely used in electrical apparatus, exhibiting low charge carrier concentration and mobility, thus low electrical conductivity, even at high fields [1-5]. Carriers in insulating polymers are introduced by impurities and contaminants, as well as by charges introduced from the electrode-insulation interface. When the electrical field applied to the polymer is higher than the threshold for space charge accumulation, charge would be introduced into the bulk material from the interface of polymer-electrode and accumulate, forming homocharge or hetero-charge [6-9]. Based on this traditional conduction mechanism, the current is a continuous flow of carriers, which gradually approaches a steady state value at a given time after the onset of voltage application.

In recent times, a new phenomenon of charge transport was revealed in some insulating polymers at relatively low electrical fields, through the use of an ultra-fast space charge acquisition and a conduction current measurement apparatus endowed of low-pass filter [10]. Charge pulses are observed to travel very rapidly through the insulation bulk with mobility 4 to 5 orders of magnitude higher than that for conventional conduction. These repetitive charge packets have magnitudes that are almost inde-
dependent of field and temperature and do not show appreciable pulse distortion and attenuation during transit, thus behaving as solitons. Such a mechanism should be, therefore, different from that responsible for traditional charge transportation by hopping through localized states (traps). Previous research work has indicated that this new conduction mechanism could be associated with the electro-mechanical compression of the polymer and polarization mode. It is known that the morphology of a polymer can be modified by nano-fillers, due to the high aspect ratio, so that the mechanical, thermal, and electrical properties of the polymer could be affected significantly by the introduction of nano-fillers [11,12].

Here, we report evidence for the effects of nano-size fillers on space charge and breakdown behaviors of epoxy/composites. The Al2O3 fillers are used in this study. The pulsed electro-acoustic technique is used to monitor space charge behaviors of the samples and the breakdown behaviors of epoxy/nanocomposites with different concentrations have been measured.

2. EXPERIMENTS

Room temperature curing epoxy resin (LY-556) supplied by Huntsman Advanced Materials Pvt. Ltd., India was used as the matrix system. The alumina (Al2O3) is ceramic filler used as reinforcement was supplied by Sigma-Aldrich., India. These ceramic fillers are of spherical shape and with diameter 90-120 nm. An open mold with cavity dimensions 225×225×6 mm was fabricated to test polymer composites. The fillers were preheated to 80 °C for 2 h to remove any moisture present and cooled to ambient temperature. Filler with 0.1, 0.25, 0.5 and 1.0 wt. % were stirred gently into liquid epoxy resin, in order to avoid any introduction of air bubbles. Resin filler mixture was then placed under the vacuum (760 □ mm Hg) for about 2 h to remove any entrapped air. Hardener was then added to the resin in the ratio of 1:10 and then stirred to ensure complete blending. The mixture was then poured into an open metallic mold coated with release agent and the mold was placed in a toughened glass chamber maintained at a low-vacuum level 400-450 □ mm Hg for about 1 h. Specimens were allowed to cure under room temperature and released from mold after 24 h. Cast composites plates obtained were of dimension mm. The plates were then post-cured at 50 °C for 2 h in a hot air oven.

The functionalization introduction by reactive groups onto the alumina surfaces was achieved by the reaction of silane with the hydroxyl groups of the alumina. A typical example can be given as follows. 2.0 g alumina particles and 2.0 g KH 570 in 100 ml of 95% alcohol solution were charged into a 300 ml flask equipped with a reflux condenser. The mixture was refluxed at the boiling temperature of the solution over different times of stirring. After that, the alumina was centrifuged, and the precipitate was extracted with alcohol for 16 h to remove the excess silane absorbed on the alumina. Then the treated alumina was air-dried and allowed to react at 80 °C under vacuum for 24 h. The content of the double bonds introduced onto the alumina surfaces by the above treatment [12].

Composition of the test specimens was varied with 0, 0.1, 0.5 and 1.0 wt. % of treated and untreated alumina filler. Extreme care has been taken to avoid any undesirable filler settling effect by casting the slurry just prior to its gelling stage, all time keeping it in a stirred condition. This was done to ensure the uniform composition of cast specimens across its volume. Higher Al2O3 content means higher viscosity. Test samples were prepared from the cast composites using the diamond-tipped cutter.

The Pulsed electro-acoustic (PEA) system was used for measuring the space charge accumulation in epoxy composites. A voltage of 6.0 kV had been applied to specimens with approximately 0.2 mm thickness (The applied field is 30 kV/mm) at room temperature (approximately 28 °C) for 60 minutes. In the present case, the results from “volts-off” measurements were used to investigate charge accumulation in the specimens.

3. RESULTS AND DISCUSSION

The TEM micrograph of the 0.5 wt.% epoxy/Al2O3 composites shown in Fig. 1. The microstructure of the composite shows the complex networking of Al2O3 fillers in epoxy matrix. The inset of the Fig. 1 shows the higher magnification of the Al2O3 particles, indicating the high-quality dispersion of the fillers within the epoxy matrix.

The space charge accumulation results are shown in Fig. 2(a-f). It can be seen that, how the overall distribution of space charge in nanocomposites evolves with time during charging and discharging at room temperature. During the charging process, positive charge injection occurs from the anode, as can clearly be seen in Fig. 2. On short circuiting after voltage application, small amounts of positive and negative charge are initially evident near the anode and the cathode, respectively, but these so-called homo-space charges, decay gradually with time at this temperature. The unreinforced epoxy, charges are dictated by homo-charges, which means that the charge injection takes place from both electrodes. Compared with the unreinforced samples, it is noticed that composites has a slower initial charge build-up rate and the magnitude is less than that in the unreinforced epoxy.

The characteristic variation in short time break down voltage of epoxy/Al2O3 nanocomposites with different weight percentage of pristine and silane modified Al2O3 under high applied voltage is shown in Fig. 3. With an increase in weight percent of pristine and silane-modified Al2O3 in epoxy; the breakdown voltage increases up to critical concentration and remains steady thereafter. The improvement in breakdown voltage is due to the dispersion of nano-size alumina particle which depicts higher surface area resulting in altered matrix behavior.

As a result of increase in surface area of alumina nanoparticles, the interfacial area increases significantly. This leads to the increase in the volume of epoxy surrounding the alumina nanoparticles which is referred as high interaction zone. This interaction zone is much higher compared to the micron-size particle filled composites which alter the functional properties of nanocomposites. It was found that because of high packing density, the accumulated charge was reduced and threshold field at any point is reduced and the space charge distribution is altered. However, when the material is packed with nanoparticles, the fillers act as a scattering site and electrons cannot gain momentum to involve in the process of breakdown. Under the application of high voltage, the ejected electrons are accelerated by the applied field and collide directly with the highly dispersed surrounding nanoparticles and lose their momentum. It requires additional voltage to cause any catastrophic failure of insulation and cause damage to the insulation, whereas, due to these aforementioned reasons epoxy/silane modified Al2O3 nanocomposites has improved breakdown strength compared to the epoxy/pristine Al2O3 under high applied voltage. In addition, the presence of nanoparticles, decrease the erosion depth and restrict the tree formation which improves the breakdown voltage. The fractography study through scanning electron micrograph (SEM) of epoxy/alumina nanocomposite reveals a tree type pattern as shown.

To investigate the electrical insulating properties of the composites, dielectric breakdown measurements were performed in
a liquid nitrogen bath \((T = -196 \degree C)\) and the failure distributions are presented in Fig. 4. For the data analysis a two parameter Weibull model was employed. Presence of pristine and silane modified \(\text{Al}_2\text{O}_3\) reinforcements and their interactions with the matrix in composite materials cause the low dielectric losses and changes in local electric field distribution, improving the electrical insulation properties. Composite materials with such variant temperature dependence in the conductivity/insulating properties could be used in the design of devices and sensors operating in broad temperature windows.

4. CONCLUSIONS

Both the space charge accumulation and electrical breakdown

behaviors in epoxy composites loaded with different concentration of \(\text{Al}_2\text{O}_3\) fillers have been investigated using the PEA method and AC breakdown kits. The following conclusion may be drawn based on this study.

The presence of \(\text{Al}_2\text{O}_3\) particles leads to space charge accumulation inside the bulk of sample. This might be due to the presence of \(\text{Al}_2\text{O}_3\) particles modified the space charge trap distributions, especially at higher loading concentration. The test results also shown that epoxy/ \(\text{Al}_2\text{O}_3\) composites samples seems accumulate less amount of charge compare with the unreinforced epoxy.

The presents of untreated fillers show a reduction in the breakdown strength of epoxy resin composites. The \(\text{Al}_2\text{O}_3\) fillers with surface treatment, on the other hand, improved both the breakdown strength and consistence of the epoxy resin composites. The Weibull distribution plots also show that the best mixing ration for both nanocomposites with and without surface treatments is around 0.5 wt.% of filler content composites.

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REFERENCES


