IMPROVING INSULATING PROPERTIES OF EPOXY POLYMER BY A TRACE OF FULLERENE PARTICLES

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ABSTRACT

Pursuing better electrical insulating properties is an everlasting aim for insulating materials. In this work, epoxy composites filled with a trace of fullerene particles (C_{60}, 1-960 ppm) were prepared. Very low C_{60} loadings (10-100 ppm) were found to effectively improve the volume resistivity and breakdown strength. The C_{60} particles, unlike the traditional ceramic particles (e.g., alumina, silica, magnesia), can take effect at extremely low filler loading in epoxy, and therefore did not bring about any thickening effects, which was favorable to material processing. Thermally stimulated depolarization current results confirmed that C_{60} particles introduced deep-level traps in epoxy, which could be responsible for the improved insulating properties. The deep traps may originate from the high electron affinity of C_{60} particles.

1 INTRODUCTION

Since epoxy polymers show excellent electrical insulating and mechanical properties, low cost and good processability etc., they can serve as insulating materials everywhere. In order to ensure high operating reliability, prolong service life, reduce dimensions, weight as well as cost of these electronic/electrical devices, to further improve the insulating properties of epoxy-based materials is quite necessary. Nanosized inorganic ceramic fillers are introduced to the epoxy or other polymers to further tune their insulating properties, including permittivity, loss tan δ, electrical conductivity, space charge accumulation, dielectric breakdown strength, partial discharge resistance, tracking resistance, treeing life and the like [1,2]. However, even at typical loadings (1~10wt.%), due to the huge specific surface area of nanofillers, the mixtures of polymer with nanofillers often become very thick, lose their flowability more or less and thus cause big issues in manufacture of nanocomposites [3,4]. Therefore, it is very necessary to find the new fillers with highly-efficient charge-capture ability (corresponding to low filler loading) for polymer insulation.

In this work, we found ppm order of magnitude of fullerene (C_{60}) additive can increase volume resistivity and breakdown strength of a highly-insulated epoxy polymer, without any thickening effect to the uncured epoxy mixture. The micro-mechanisms were discussed based on the TSDC results.

2 RESELTS AND DISCUSSION

Fig. 1a shows the filler loading effect on the volume resistivity (ρ_v) of samples tested at room temperature. Since the epoxy polymer used in this work is specially designed for highly insulating applications, it has very high ρ_v value (7.5×10^{16} Ω·cm), compared to common epoxy systems. It is interesting to note that after addition of very small amount of C_{60} particles, the ρ_v values of nanocomposites are generally higher than that of the neat epoxy polymer, even though the C_{60} particles are traditionally considered as semi-conductive materials (C_{60} has the carrier mobility in the order of

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10^{-2} cm^2/V^{-1}s^{-1} for [5], much larger than that of 10^{-12} cm^2/V^{-1}s^{-1} for epoxy [6].). The highest ρ_v value (9.8×10^{16} Ω·cm) is found for the composite sample containing only 24 ppm C_{60} particles, i.e. ~23% increase in ρ_v, compared to that of the neat epoxy polymer. With further increasing the C_{60} particles loading (>480 ppm), the ρ_v value falls below that of the neat polymer. Fig. 1b shows the temperature dependence of ρ_v of some selected samples. The ρ_v values of samples are found to decrease gradually with the temperature. From 293 to 348 K, the ρ_v value of the neat epoxy declines by 66%, which is because increasing temperature allows more carriers (mainly composed of electrons and anions for epoxy [7]) of epoxy polymer to come out of the traps and thus producing higher current values. Comparatively, the sample containing 24 ppm C_{60} particles presents higher ρ_v values than the neat epoxy polymer in the whole test temperature range, while the sample containing 480 ppm C_{60} particles shows, at different temperatures, almost similar ρ_v values compared to the neat epoxy polymer.

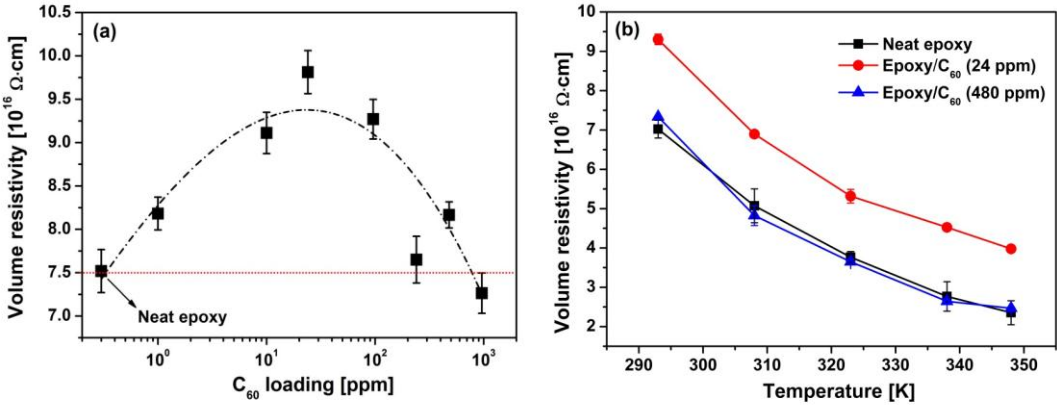


Fig. 1: The volume resistivity of epoxy and epoxy/C_{60} composites as a function of the C_{60} particle loading, tested at (a) room temperature (293 K), (b) elevated temperature (293-348 K).

The breakdown strength of a material is defined as the maximum electric field intensity that material can withstand without causing it to breakdown. Breakdown of material forms an electrically conductive path and a disruptive discharge through the material, which destroys the insulating capability [4]. Fig. 2 displays the DC breakdown strength of the samples as a function of filler loading. The specific breakdown strength, E_0, has the highest value for the sample with 100 ppm C_{60} particle, i.e. 536 kV/mm, ~41% improvement compared to that of the neat epoxy (380 kV/mm).

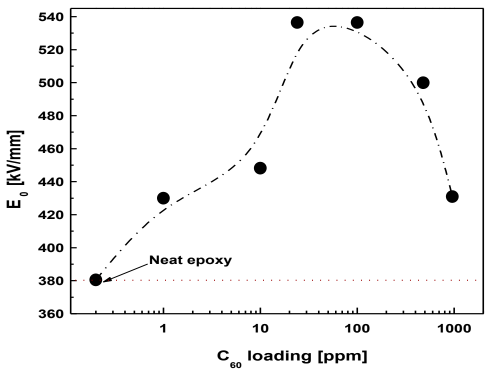
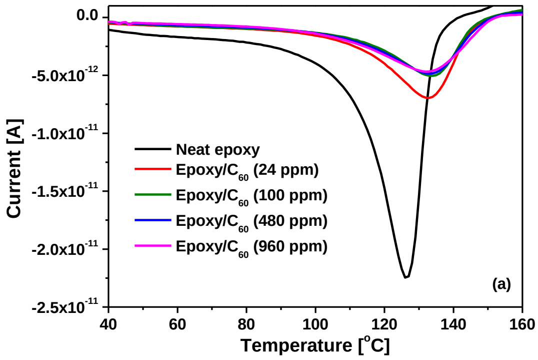


Fig. 2: DC breakdown strength of epoxy and epoxy/C_{60} composite samples.

TSDC technique is usually used in polymer dielectrics to characterize trap properties, providing insight into the internal micro-mechanisms behind electrical insulating phenomena. Fig. 3a shows the TSDC spectra for neat epoxy and epoxy/C_{60} composites. Two important parameters: the temperature at peak current (T_p) and the released charge by thermal activation (Q_r, calculated by integrating the area under current-time curve for TSDC test), can be obtained from this figure. Fig. 3b shows that the addition of a small amount of C_{60} particles (24-960 ppm) increases T_p of epoxy polymer slightly from ~126 to ~133 ℃, which means the epoxy/C_{60} composites, compared to the neat epoxy, need more energy to release the charge carriers trapped, i.e. C_{60} particles can restrict mobility of charge carriers, possibly due to their high electron affinity of ~2.7 eV [8-10]. It is established the unique three-dimentional structures make fullerene have good electron acceptability of up to six electrons [9]. Recent work suggested that the electronic structure of functional groups on nanoparticles can indeed control the dielectric properites of the epoxy composites; the nano-TiO_2 particles modified by electron-withdrawing groups (i.e. Ph-Cl, Ph-NO_2 with the electron affinity of ~1-1.2 eV [10]) offered epoxy polymer significant improvements in dielectric breakdown strength, reduction in leakage current and dielectric loss, compared to the unmodified ones. Although it is impossible to compare our samples with the samples in above work directly, it is noted that the C_{60} particles are also an electrophilic material with high electron affinity, therefore, it is reasonable that the deep-level traps originate from the high electron affinity of C_{60}.

On the other hand, the addition of C_{60} particles reduces significantly Q_r from ~3.1×10^{-8} C for the neat epoxy to ~1.1×10^{-8} C for the C_{60}-filled composites. Since Q_r originates from the relaxation of epoxy dipole orientation and the release of space charges injected from electrodes [11], this result may suggest that C_{60} particles can reduce the level of epoxy dipole orientation and also suppress space charge injection/accumulation.



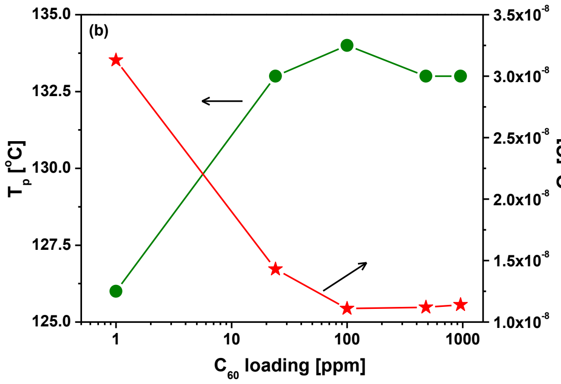


Fig. 3: (a) TSDC spectra of neat epoxy and epoxy/C_{60} composites, and (b) the peak temperature (T_p) and released charges (Q_r) as a function of the C_{60} particle loading.

3 CONCLUSIONS

The effects of C_{60} particle loading on the insulating properties of epoxy were investigated. A very small amount of C_{60} particles were found to increase the volume resistivity of epoxy and the DC breakdown strength simultaneously. According to TSDC results, C_{60} particles generated deep-level traps in epoxy, which was responsible for the improved insulating properties; the deep-level traps may correlate with the high electron affinity of the C_{60} particles. The small amount of C_{60} particles reduced the viscosity of uncured epoxy resin, this is definitely beneficial to material process. It is speculated that this phenomenon would be universal for other electrophilic carbon materials and may open up a new way to modify the insulating polymers.

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