EPOXY NANOCOMPOSITES CONTAINING CERAMIC FILLERS FOR ELECTRICAL APPLICATIONS

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Introduction

High performance epoxy nanocomposites are emerging as a class of materials for their wide industrial applications as structural composites, adhesives, surface coating and as insulating material in electrical motors. The insulation of rotating machines is subjected to multiple and concomitant stresses during operation, e.g. electrical, mechanical and thermal [1]. The generated heat, if not dissipated, might potentially affect the structural integrity of the final product, thus it is necessary to improve thermal conductivity of dielectric insulators. Unfortunately, the relatively disordered polymeric structure of epoxy resins is also responsible of very poor thermal conductivity [2]. The use of epoxy nanocomposites as dielectrics requires optimal dielectric properties, therefore in this direction the addition of insulating ceramic nanoparticles to epoxy resin can provide a promising material with good dielectric and thermal properties [3].

Experimental

Materials

A diglicydyl ether of bisphenol-A epoxy resin with an epoxide equivalent of 187g/equiv. cured with methyl tetrahydrophthalic anhydride hardener supplied by Elantas Camattini S.p.a. was used for the investigations. The addition of 0.25 phr of methyl imidazole (supplied by Elantas Camattini S.p.a.) catalysed the crosslinking reaction. Alumina nanoparticles with an average particle sizes smaller than 50 nm (Al_2O_3, supplied by Sigma Aldrich), silica (SiO_2, Aerosil® 200) and titanium dioxide (TiO_2, Aeroxide P25) produced by Degussa with average particle of 12 and 21nm respectively, were used for composites preparation. The amount of ceramic particles added was 1 and 3 weight percent. Combined dispersion techniques have been used for mixture preparation, namely mechanical mixing and sonication [4]. The nanoparticles have been added to anhydride hardener and then mechanically mixed with a high speed homogeneizer "*Turrax T25*" at 6500 rpm for 10 min. The mixture has been sonicated in an ultrasonic processor *"Hielscher UP200S"* with a 20% of maximum amplitude (200W) for about 30 min. After addition of the suitable amount of EC01, the system has been further sonicated for 10 min. Finally, the samples has been cured according to the thermal cycle suggested by technical data sheet (4h at 80°C, 3h at 100°C and 4h at 140°C).

Apparatus and Procedures

A scanning electron microscope (SEM, type Leica S440) was used to analyze the inorganic filler dispersion.

The thermal conductivity of neat epoxy resin and nanocomposites has been estimated by modulated DSC (MDSC), according to ASTM E 1952 .

Dynamic-mechanical analysis was carried out by a TA Instruments DMA 2980 at a heating rate of 5°C/min in a range of temperature from 0 to 200°C. Single cantilever geometry has been adopted, with a deformation amplitude of 15μm at the frequency of 1 Hz.

The following electrical properties were evaluated: relative dielectric permittivity (ε_r), dissipation factor (tan δ) and dc volume resistivity (ρ_v). Permittivity and tan δ have been measured in the frequency range of 10^2-10^6 Hz by using an impedence analyzer HP 4192A.

The volume resistivity has been measured at 20°C, by holding the specimens in a suitable shielded cell and using a stabilised dc source and a picoammeter HP 4140B.

The data presented in this paper are an average value of 5 samples.

Results and Discussion

DMA. Addition of ceramic particles determines, as expected, an increase in elastic modulus from the value of ~2.7 GPa observed for the unfilled resin, reaching the maximum value of ~3.25 GPa for the composite containing 3% of TiO_2 (fig.1).

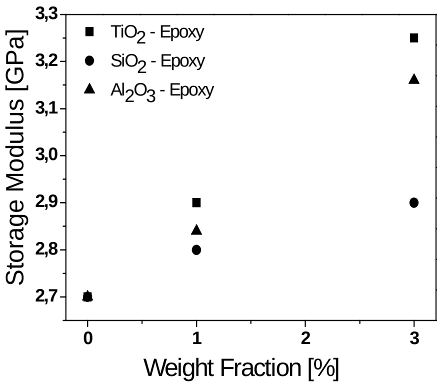

Figure 1. Storage Modulus of epoxy nanocomposites.

The glass transition temperature of composites, reported in table 1 increases of about 7°C for sample containing 3%wt of SiO_2 filler. The addition of rigid nanoparticles with a very large surface area and the formation of the interface boundary region between matrix and solid particles reduces the polymer chains mobility resulting in an increased glass transition temperature.

Table 1 T_g values assessed by DMA analyses for the nanocomposites and the neat epoxy resin.

Filler content [%wt]	Al_2O_3	SiO_2	TiO_2
0	139	139	139
1	138	145	141
3	145	147	144

DSC. The thermal conductivity of neat epoxy and its various hybrids has been evaluated by modulated DSC method. At lower filler content, the thermal conductivity remains approximately constant for all specimens containing different fillers. Increasing Al_2O_3 content up to 30%wt, the λ value is roughly doubled compared to the neat resin. This improvement, from 0.19 W/mK, for neat resin, to 0.29 W/mK for 30% Al_2O_3 sample, is due to the formation of a percolative pathway for heat conduction.

Electrical tests. dc volume resistivity has been measured by applying a dc voltage of 1400 V on thin samples (thickness ranging between 0.6 and 0.8 mm), corresponding to an applied electrical field ranging from 15.5 to 23.0 kV/cm. The values of dielectric current have been recorded after 120 s from the application of the dc voltage. In Figure 2 dc resistivity ρ_v vs. the filler content % wt is reported for the tree different typologies of samples. It can be noted a decrease of ρ_v with respect to the pure epoxy resin, which is more evident in the TiO_2 samples (in the 3% wt samples the resistivity is one order of magnitude lower with respect to pure epoxy resin).

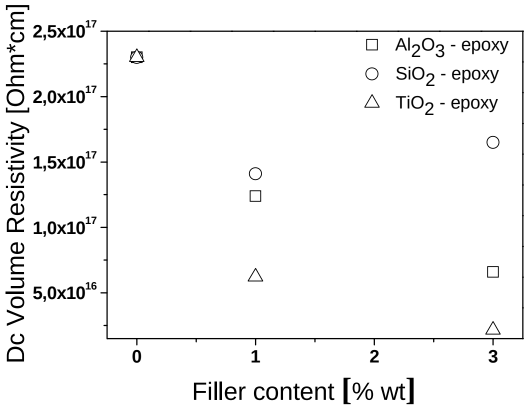

Figure 2. dc volume resistivity of epoxy nanocomposites.

Conclusion

The paper presents results on the investigation of epoxy-nanocomposites containing ceramic nanoparticles. At low filler content (1% and 3%wt), the threshold of continuous pathway is not reached, and thermal conductivity λ values are only slightly reduced with respect to neat matrix. At high filler content (30%wt) the formation of a percolative heat sink pathway increases thermal conductivity.

The elastic moduli and the T_g of the composites increased compared with neat resin. A slight reduction in the epoxy dc volume resistivity is observed by incorporating a small amount of nano-filler into epoxy matrix.

References

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