Development of Poly(dimethylsiloxane)/BaTiO₃ Nanocomposites as Dielectric Material

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Abstract. Polymer-ceramic nanocomposites with controlled dielectric properties are prepared using poly(dimethylsiloxane) elastomer as base matrix and barium titanate as filler. Barium titanate (BaTiO3) used in this study is prepared by solid state reaction at high temperature. The effect of BaTiO3 nanoparticles on electrical and mechanical properties are extensively studied and found that dielectric constant of nanocomposites increases significantly with the increase in BaTiO3 concentration where as volume resistivity decreases continuously. Different mechanical properties are also studied for all the composites in order to find the effect of filler concentration. Morphology of the prepared BaTiO3 was studied by field emission scanning electron microscope (FESEM).

Introduction

Polymer nanocomposites (nanoparticles dispersed in a polymer matrix) are of great interest because addition of nanoparticles to a polymer matrix can enhance mechanical, thermal, barrier, and other properties [1]. Most of these composites have either, nanoclay, nano fibre, or carbon nanotube dispersed in appropriate polymer matrix [2]. These composites have drawn great interest for its versatile applications in the field of electronic materials [3, 4] such as integrated decoupling capacitors, acoustic emission sensors, electronic packaging materials [4–6] and angular acceleration accelerometers [7, 8]. Many studies have been done on BaTiO₃ due to their remarkable optical and electronic properties [9].

In the present study, polydimethylsiloxane (PDMS) elastomer and prepared barium titanate ($BaTiO_3$) nanoparticles were used to develop polymer-ceramic nanocomposites. Different electrical and mechanical properties of these composites were studied at various filler concentration. The influence of the filler on electrical and mechanical properties of these nanocomposites was also measured.

Experimental section

Materials used. Polydimethylsiloxane elastomer was purchased from D J Silicone: Shore A hardness=40 and density=1.12 g/cc. Titanium dioxide and Ba(OH)₂·8H₂O were procured from Merck chemicals, India. Dicumyl peroxide, MP=80°C, purity=98% (Sigma-Aldrich chemical company, USA) was used for curing purpose.

Synthesis of nano barium titanate (BaTiO₃) by solid state reaction. Barium titanate nano particles were prepared from $Ba(OH)_2 \cdot 8H_2O$ and TiO_2 by high temperature solid state reaction. Requisite amounts of two oxides were mixed intimately. The mixture was then subjected to sintering at high temperatures (600°C-1100°C). Finally, the product formed was powdered and used for further investigation.

Preparation of nanocomposites and samples. Mixing of $BaTiO_3$ and other ingredients into polymer matrix (PDMS) were done using an internal mixer (Brabender–plasti corder) for 5 minutes. All the nanocomposites were prepared by taking the concentration of curing agent same and only varying the concentration of the filler (0 php, 30 php, 50 php, and 70 php) in polymer matrix. Then these compounds were passed through a two roll mill to make them into sheet form. In these formulations, all ingredients were taken as parts by weight per hundred parts by weight of polymer (php). Monsanto rheometer R 100 was used to determine the optimum cure time at 150°C for 45 minute for these composites. Moulding was done at 150°C in an electrically heated press for a specified time (obtained through Monsanto Rheometer R 100) at a pressure of 5 Mpa.

Characterization. The DC volume resistivity of neat elastomer and their nanocomposites were measured using high resistivity meter (model. Agilent 4339B). The dielectric properties of these nanocomposites were measured using precision LCR meter (model. Quad Tech 7600). Tensile test was carried out according to the ASTM D 412 specifications using Hounsfield tensometer (model. H10KS). The morphology of prepared BaTiO₃ was studied using field emission scanning electron microscope (FESEM, model. SUPRA 40, Carl Zeiss SMT AG, Germany).

Results and discussion

Electrical and mechanical properties. Log f vs. dielectric constant (ε ') and dielectric loss (ε ") plots for all the nanocomposites are presented in Fig. 1a and 1b respectively. It is observed from these figures that dielectric constant (ε ') is significantly increased with filler loading and is more pronounced at low frequency region. The significant increase in dielectric constant especially at low frequency region is mainly due to increase in the interfacial polarization. Similarly, dielectric loss is also increased with increase in filler concentration (Fig. 1b), but the increase is very less as compared to composites containing conventional carbon fillers (carbon black, carbon fibre, and carbon nanotubes etc.). Composites containing carbon fillers shows more dielectric loss due to their highly conductive nature even sometimes it exceeds dielectric constant value.



Fig. 1 Log f vs. ϵ' (a), log f vs. ϵ'' (b) plots for PDMS/BaTiO₃ composites

The DC resistivity of composites is decreased with increase in filler loading as shown in Fig. 2. This is mainly because of the fact that DC resistivity of particulate filler is lesser than the polymer matrix. In addition, particulate filler often contains some amount of moisture, which also affects the DC resistivity of the composites. Fig. 3 represents the effect of filler (BaTiO₃) loading on tensile strength and % elongation at break (% E.B.). From this figure, it is observed that both tensile strength and % E.B. decreased continuously with the increase in filler concentration. The decrease in both tensile strength and % E.B. with filler loading are due to non-reinforcing nature of BaTiO₃ in PDMS matrix. Fig. 4a and 4b represents the FESEM images of the prepared nano barium titanate (BaTiO₃). From this image, it is observed that barium titanate have bamboo leaf like structure.



Fig. 2 Effect of filler loading on DC resistivity



Fig. 3 Plots of tensile strength and % E.B. against the filler concentration



Fig. 4 FESEM image of nano BaTiO₃ powder

Conclusions

Graded dielectric material with varying dielectric properties can be made from PDMS-BaTiO₃ nanocomposites by changing the concentration of nano barium titante. The addition of barium titante to polydimethylsiloxane (PDMS) elastomer increased both dielectric constant and loss factor of the resultant nanocomposites. Barium titante is non-reinforcing filler for PDMS matrix. Barium titante particles have bamboo leaf like shape as apparent from FESEM image.

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