

Poly(dimethylsiloxane)-TiO₂ Nanocomposites for Dielectric and Piezoelectric Applications

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Abstract—Flexible polydimethylsiloxane-titania (PDMS-TiO₂) nanocomposites of different composition are prepared using two mixing methods/techniques. In the present study, PDMS elastomer is used as base matrix and titania (TiO₂) as ceramic filler. The effect of titania on thermal, electrical, and mechanical properties is investigated for different composites at various concentration of titania. A variety of electrical and mechanical tests are performed on the resultant composites and found that dielectric constant of composites increased significantly with the increase in titania concentration whereas volume resistivity of composites decreased. The effect of titania concentration is also studied with respect to tensile strength, % elongation at break (% E. B.), and tensile modulus. The effect of pressure on dielectric constant and resistivity of composites has also been studied in order to determine the piezoelectric behavior of the composites. Filler dispersion/distribution in PDMS matrix is studied through field emission scanning electron microscopy (FESEM) and high resolution transmission electron microscopy (HRTEM).

Keywords—poly(dimethylsiloxane), dielectric properties, mechanical properties, nanocomposites, titania

1. Introduction

Polymer composites are promising candidates for embedded capacitor as these materials have high dielectric constant, low dielectric loss, and good compatibility with printed-circuit-boards (PCB) [1]. In the present investigation, titania particles are used in order to study its effect on thermal, electrical, and piezoelectric properties of PDMS-titania composites. Composites derived from inorganic fillers dispersed in polymer matrix have received significant attention because of their modified mechanical properties coupled with tailor made dielectric properties and in order to prepare suitable dielectric materials [2, 3]. Polymer-ceramic composites have different

applications in the field of electronics [4, 5] such as acoustic emission sensors, angular acceleration accelerometers [6] and electronic packaging materials [4]. Dielectric materials are capable of storing large amounts of electrical energy which are desirable for many electronic and power devices. Polymers are seldom used in their pure form and are usually combined with mineral fillers [2]. These fillers are used with polymer in order to improve mechanical, thermal, electrical properties, and dimensional-stability of the final composites. Titanium dioxide is an inorganic filler having large permittivity, so used for insulating material in electronics and dielectric for capacitors [7]. Tailor made dielectric material/capacitor with controlled dielectric properties can also be obtained using PDMS as matrix filled with electro-ceramics like titania (TiO₂), BaTiO₃, and PZT etc having high dielectric constant [8-10].

In the present study, polydimethylsiloxane (PDMS) elastomer and titania particles are used to prepare polymer-ceramic composites. Different electrical and mechanical properties of these composites were studied and found that dielectric constant of these composites was increased significantly with the addition of filler, whereas volume resistivity was decreased. The influence of titania particles on dielectric and piezoelectric properties of these composites was also measured.

2. Experimental Section

2.1. Materials Used

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

2.2. Preparation of Nanocomposites and Samples

Mixing of titania filler and other ingredients into PDMS matrix was done in an internal mixer (Brabender-plasti corder) for 10 minutes. All the nanocomposites were prepared by taking the concentration of curing agent same and only varying the concentration of the filler (0 php/0 wt %, 30 php/23.08 wt %, 50 php/33.34 wt %, and 70 php/41.18 wt %) in polymer matrix. Then the mixed mass was passed through a two roll mill to make them into sheet form. In these formulations, all ingredients were taken as parts by weight per

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hundred parts by weight of polymer (php). Rubber process analyser (RPA) 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 10 min as obtained from RPA at a pressure of 7 Mpa.

3. Characterization

The DC volume resistivity of PDMS elastomer and its composites was measured using high resistivity meter (model. Agilent 4339B). The normal and pressure dependent dielectric properties of these composites 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 PDMS-titania composites was studied using field emission scanning electron microscope (FESEM, model. SUPRA 40, Carl Zeiss SMT AG, Germany) and high resolution transmission electron microscope (HRTEM, Model JEM-2100). Thermogravimetric analysis was carried out in TA Instruments (model Q 50), at a heating rate of 20°C/min under nitrogen atmosphere up to 650°C.

4. Results and Discussion

4.1. Electrical Properties

The DC volume resistivity of pristine PDMS and its composites is decreased with increase in filler loading as shown in Fig. 1. The decrease in resistivity with titania loading is due to the lower resistivity of titania particles compared to the pristine PDMS. Moreover, inorganic particles/oxides often contain moisture on their surface, which also affects the resistivity of the composites.

The plot of log f vs. dielectric constant (ϵ') for all the composites is presented in Fig. 2. It is observed that there is increase in dielectric constant (ϵ') with filler loading at all frequency. The increase in dielectric constant with titania loading is also frequency dependent and is more pronounced at low frequency region. The high dielectric constant at low frequency region is mainly due to the contribution of interfacial polarization.

The effect of external stress/pressure on dielectric constant of composites at 1 kHz is presented in Fig. 3. It is observed that dielectric constant of composites is increased with the increase in pressure. The change in dielectric constant with pressure is also composition dependent as apparent from the figure. The change in dielectric constant with external stress is more for composite containing higher titania loading. There is change in geometry of the interface by the application of external stress as a result there is increase in dielectric constant of the composites. Moreover, there is increase in charge density of the system by the application of pressure. The increase in charge density leads to the increase in dielectric constant of the system. The effect of stress on DC

resistivity is also studied and found that resistivity of composites is decreased continuously with increase in external stress (figure not given).

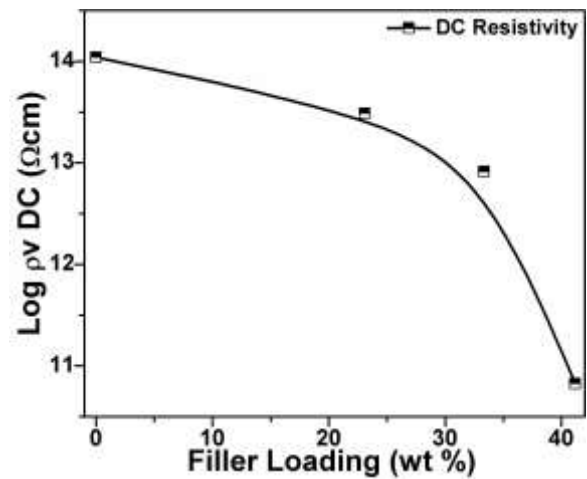


Fig. 1 Effect of titania loading on DC volume resistivity.

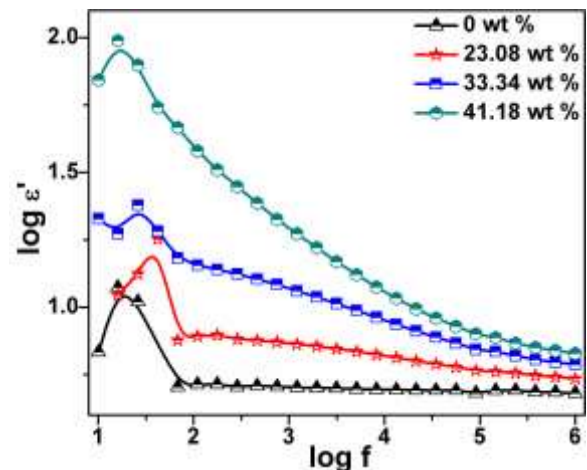


Fig. 2 Log f vs. ϵ' plot for PDMS-TiO₂ composites.

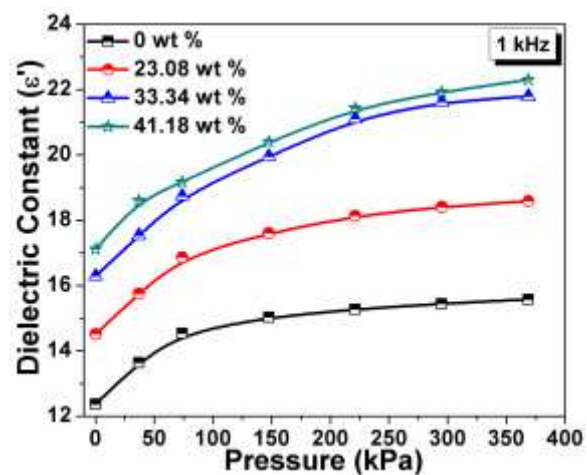


Fig. 3 Effect of external stress/pressure on dielectric constant of composites at 1 kHz.

4.2. Mechanical Properties

Fig. 4 represents the effect of titania filler loading on both tensile strength and % elongation at break (% E. B.). It is observed that tensile strength and % E. B. decreased with the increase in titania loading in PDMS matrix. The decrease in both tensile strength and % E. B. with filler loading is due to the non-reinforcing nature of titania particles for PDMS matrix. The variation of tensile modulus with titania loading in PDMS matrix is presented in Fig. 5. Both tensile modulus at 100 % and 200% elongation is found to increase with the increase in titania loading.

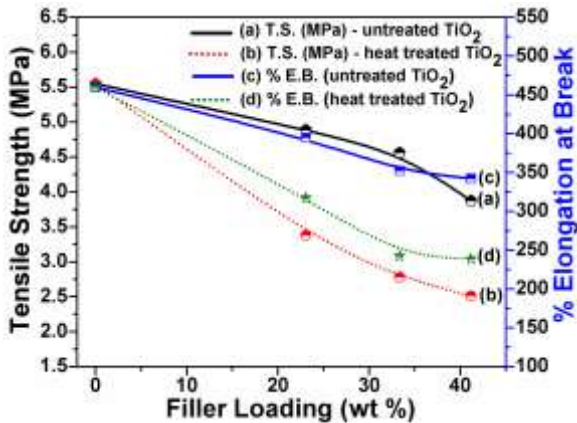


Fig. 4 Effect of titania loading on tensile strength and % E. B.

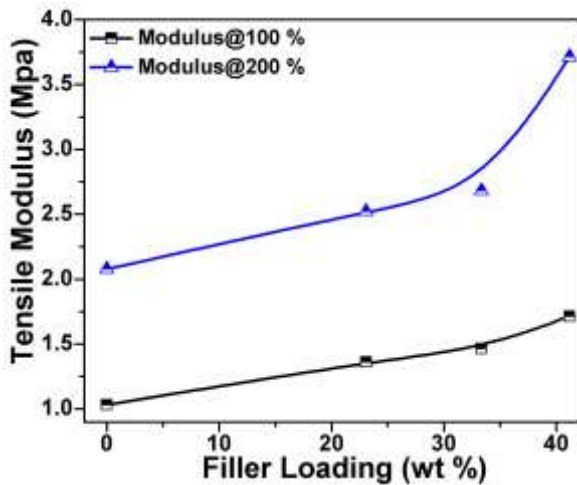


Fig. 5 Effect of titania loading on tensile modulus.

4.3. Morphological Analysis

The dispersion and distribution of titania particles in PDMS matrix is studied through FESEM and HRTEM. The FESEM images of PDMS-titania composites containing different concentration of titania (23.08 wt % and 41.18 wt %) are presented in Fig. 6a-b. It is observed that filler particles are well dispersed in the PDMS matrix both at low and high loading. It is clear from these figures that at lower loading,

filler particles are properly wetted by the PDMS matrix. However, wettability of filler particles by PDMS matrix is reduced with the increase in filler loading because of dilution effect. The dispersion and distribution of titania particles in

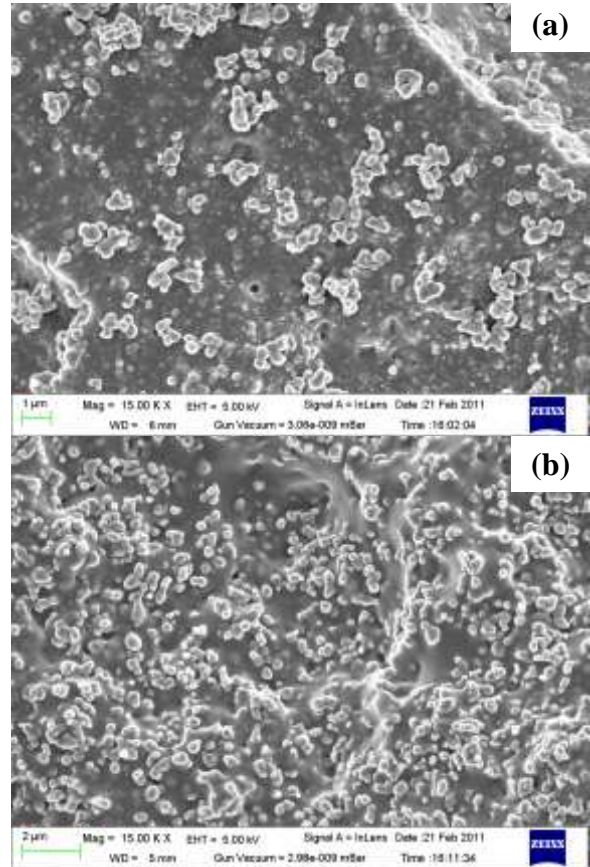


Fig. 6 FESEM image of PDMS-titania composite containing (a) 23.08 wt % and (b) 41.18 wt % of TiO₂.

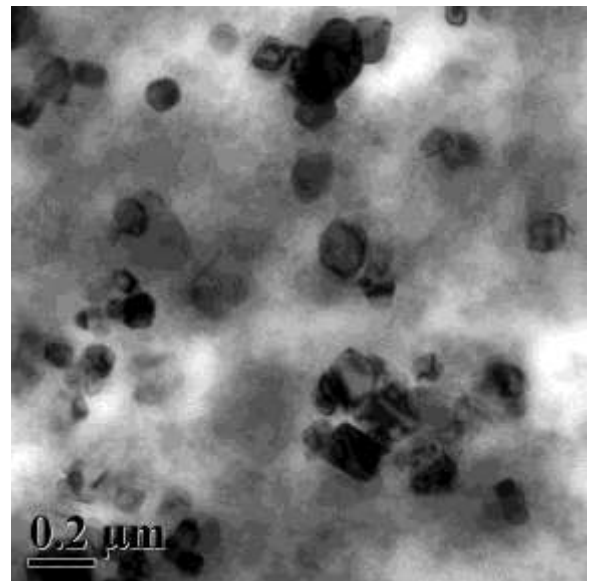


Fig. 7 HRTEM image of PDMS-titania composite containing 23.08 wt % TiO₂.

PDMS matrix is also understood from HRTEM image (Fig. 7). It is observed that titania particles are well distributed in the PDMS matrix with little agglomeration.

4.4. Thermogravimetric Analysis

Thermo gravimetric analysis (TGA) in nitrogen atmosphere was conducted at a heating rate of 20°C/min for pristine PDMS and its nanocomposites. Fig. 8 represents the loss in weight of PDMS-titania composites with the increase in temperature. It is observed that there is increase in thermal stability of composites with the increase in titania loading in PDMS matrix as the thermal stability of titania particles is more than the PDMS matrix.

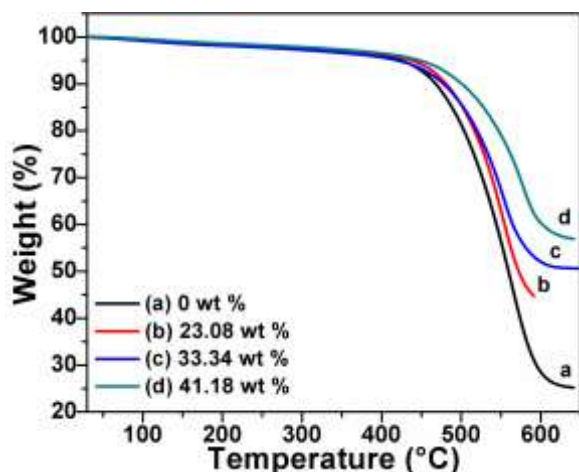


Fig. 8 Thermogravimetric analysis of PDMS-titania composites.

5. Conclusions

Flexible dielectric material with controlled dielectric and piezoelectric properties can be obtained from PDMS-TiO₂ composites by changing the concentration of titania particles in PDMS matrix. The dielectric constant of composites is increased with the filler loading whereas volume resistivity decreased. Titania is a non-reinforcing filler for PDMS matrix. Mechanical properties like tensile strength and % elongation at break (% E. B.) are decreased with the increase in titania loading but modulus is increased with the loading. There is increase in dielectric constant and DC conductivity with the increase in external pressure which is also dependent on the composition.

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