

Electrical, Dielectric, and Electromagnetic Shielding Properties of Polypropylene-Graphite Composites

Varij Panwar,¹ Jong-Oh Park,¹ Suk-Ho Park,¹ Sanjeev Kumar,² R. M. Mehra²

¹School of Mechanical and Systems Engineering, Chonnam National University, Gwangju 500-757, Republic of Korea

²Department of Electronic Science, University of Delhi South Campus, New Delhi 110 021, India

Received 19 June 2008; accepted 11 November 2008

DOI 10.1002/app.29702

Published online 7 October 2009 in Wiley InterScience (www.interscience.wiley.com).

ABSTRACT: The conducting polymer composite material is desired to have a high dielectric constant and high dissipation factor in low and high frequency ranges, so that it can be used in charge storing devices, decoupling capacitors, and electromagnetic interference (EMI) shielding applications. Currently, on-going research is trying to enhance the dielectric constant of ceramic powder-polymer, metal powder-polymer, and nanotube-polymer composites in the low frequency region. In this article, we present the dielectric properties of polypropylene (PP)-graphite (Gr) composites in low and radio frequency ranges. Furthermore, the EMI shielding properties of these composites are examined in the radio frequency range. The PP-Gr composites were prepared by mixing and the hot compression mold technique. The electrical conductivity and dielectric constant of

PP-Gr composites with graphite volume fraction follow the power law model of percolation theory. The percolation threshold of the composites is estimated to be 0.0257 (~ 5wt % of Gr). The current of PP-Gr composites as a function of voltage shows a nearly ohmic behavior above the percolation threshold. Shore-D hardness of the composites is decreased with the addition of conducting filler. The PP-Gr composites exhibit a high dielectric constant and high dissipation factor with the addition of graphite in low frequency and radio frequency regions, so they can be used in the proposed applications. © 2009 Wiley Periodicals, Inc. *J Appl Polym Sci* 115: 1306–1314, 2010

Key words: polymer composites; polypropylene; graphite; percolation threshold; electromagnetic shielding

INTRODUCTION

Conductive polymer composites can be obtained by blending an insulating polymer matrix with conductive fillers. Graphite powder,^{1–4} carbon black,^{5–8} metallic powder,^{9,10} carbon nanotube^{11–13} and piezoelectric powder^{14,15} as the conducting filler have been used extensively, as described in the literature. Some of the advantages of using polymer and graphite to realize conducting polymer composites are less weight, resistance to corrosion, flexibility, and easy processing. Conducting polymer composites can be applied as current limiters,^{16,17} charge storage capacitors^{14,18,19} and antistatic materials for electromagnetic interference shielding^{20,21} of electronic devices.

The electrical conductivity of polymer composites can be increased with the addition of conducting filler and undergoes insulator-conductor transition at a certain filler concentration, which is known as the perco-

lation threshold. Insulator-conductor transition behavior of electrical conductivity near the percolation threshold is explained by percolation theory.^{22,23} Shen et al.²⁴ studied the structure and electrical properties of Grafted Polypropylene-Graphite nanocomposites, prepared by solution intercalation, and the percolation threshold of these nanocomposites was 0.67 vol %. Kalaitzidou et al.²⁵ studied the flexural and electrical properties of exfoliated graphite-polypropylene nanocomposites, which were prepared by the melt mixing technique, and the percolation threshold of these nanocomposites was 0.3 vol %. Zois et al.²⁶ studied the AC conductivity and dielectric constant of polypropylene/carbon black (PP/CB) composite systems, whose percolation threshold was calculated to be 6.2 wt % of CB. Dweiri and Sahari²⁷ studied the electrical properties of carbon-based polypropylene composites for bipolar plates in a polymer electrolyte membrane fuel cell (PEMFC), and these composites were fabricated by two methods, namely, melt compounding and solution blending. Composites fabricated by solution blending of PP with conductive fillers tend to have higher conductivities than melt-compounded composites.

Because of a specific property of conducting polymer composites, the dielectric constant of conducting polymer composites also increases with the addition of conducting filler. Especially, near the percolation threshold, a sharp change in the dielectric constant is

Correspondence to: J.-O. Park (jop@jnu.ac.kr) and S.-H. Park (spark@chonnam.ac.kr).

Contract grant sponsor: Korea Ministry of Knowledge Economy (Strategy Technology Development Programs); contract grant number: 10030037.

Contract grant sponsor: DRDO, Govt. of India; contract grant number: ERIP/ER/0503529/M/01/850.

observed. At the percolation threshold, many conducting particles are isolated by thin insulating layers. Therefore, near the percolation threshold, the composites can become a capacitor and therefore, can be applied in charge storing devices, and electroactive polymers (EAP) and embedded capacitor applications. Different types of percolative polymer composites have been studied for capacitor and electroactive polymers (EAP) applications. Bai et al.²⁸ studied the dielectric behavior of ceramic powder [Pb(Mg_{1/3}Nb_{2/3})O₃-PbTiO₃]-ferroelectric polymer P(VDF/TrFE) composites and found that the dielectric constant of the composites was 250. Dang et al.²⁹ investigated the dielectric behavior of three-phase MWNTs/BaTiO₃/PVDF composites and found that maximum value of the dielectric constant of the composites was 450. Choi et al.³⁰ investigated the effects of BaTiO₃ on the dielectric behavior of BaTiO₃-Ni-polymethyl methacrylate composites. Huang et al.³¹ investigated the electromechanical responses of a high dielectric constant (>1000) polymer composite material and fabricated the composite by combining conductive polyaniline particles within a poly(vinylidene fluoride-trifluoroethylene-chlorotrifluoroethylene) terpolymer matrix. Huang et al.³² studied the electromechanical responses of high dielectric constant polymer composites and fabricated them by combining the high-dielectric copper phthalocyanine oligomer (PolyCuPc) and conductive polyaniline (PANI) within the polyurethane (PU) matrix, an all-organic three-component.

A major application of conducting polymer composites is electromagnetic interference (EMI) shielding. The EMI shielding effectiveness (SE) is needed to protect electronics instrument from electromagnetic interference/radio frequency interference (EMI/RFI), which is emitted by computer circuits, radio transmitters, cellular phones, electric motors and overhead power lines. Conducting polymer composites have been widely used in the area of EMI/RFI shielding, because of their unique combined properties of electrical conduction, corrosion resistance, low density and flexibility, which are better than those of metal materials, which have severe corrosion and weight penalties that greatly degrade the SE of the shields.

In this article, composites are prepared by mixing and the hot compression mold technique. The composites consist of polypropylene (PP) as the insulating polymer matrix and graphite (Gr) as the conducting filler. Polypropylene is stronger, stiffer and more brittle than polyethylene. Polypropylene's glass transition occurs at around minus 30°C and its melting point occurs around 165°C and has a lower degree of crystallinity. Because of these specific properties of PP, PP-Gr conducting polymer composites can be used as current limiters, anti static material and EMI shielding material, and in various applications like charge storing devices and embedded capacitors. In the present

study, the electrical conductivity and current-voltage (I-V) of PP-Gr composites have been investigated for different volume fractions of Gr (ϕ). The observed insulator-conductor transition of the electrical conductivity and dielectric constant of PP-Gr composites have been explained by the power law model of percolation theory. The dielectric constant, dissipation factor and AC conductivity of these composites were analyzed as a function of low and high frequencies. The shielding effectiveness of PP-Gr composites has also been studied in the radio frequency range (1 MHz–3 GHz). Finally, the hardness of the composites has also been examined as function of the Gr content.

EXPERIMENTAL

Materials

The matrix polymer is a commercial grade (H200 MA) PP resin, in the form of granules, manufactured by Indian Petrochemicals Corp. It is a semicrystalline, white, semiopaque commodity thermoplastic made in a very wide variety of grades and modifications. The various physical parameters of the PP used in the present work are as follows: density = 0.95 g/cm³, hardness (shore D) = approximately 68–83, melt flow = 0.5–136 g/10 min, melting temperature = 165°C, molecular weight = 50,460, dielectric constant @ 1 MHz = 2.2–2.6, dissipation factor @ 1 MHz = 0.0003–0.0005 and volume resistivity = 1×10^{13} Ω-cm. The electrical conducting filler is graphite powder with the average particle size ranging from 10 to 20 μm, supplied by Graphite India. Some properties of graphite are as follows: color = black, density = 1.75 g/cm³, maximum temperature = 3600°C, modulus of elasticity (Young modulus) = 4.8 GPa, electrical resistivity = 7.5×10^{-5} Ω-cm and carbon-graphite% = 30–70.

Composite sample preparation

The PP granules were ground to obtain finer particles (212–250 μm) using the Arthur H. Thomas type Willey Grinder. The polymer and filler powders were tumble mixed thoroughly for 4 h in a glass beaker with a magnetic stirrer. The speed of magnetic stirrer was kept around 400 rpm without any heating. This process coated the conducting graphite powder on the surface of the PP particles, and is also referred to as the prelocalization of the conductive phase. Prolong mixing improves the homogeneity of the spatial distribution of the conductive particles and their uniform coating thickness on the PP particles. The tumble mixed, prelocalized powders were prepared to obtain different graphite contents in the range about 0, 1, 2, 3, 5, 7, 10, 15, 20, and 25 wt % of graphite under similar conditions. The corresponding volume fraction of

Gr (ϕ) of these samples were 0, 0.0054, 0.011, 0.015, 0.026, 0.036, 0.052, 0.075, 0.097, and 0.119.

The graphite coated PP powder was pressed initially for 5 min at room temperature to eliminate any void formation within the pellet using a hydraulic press (Shimadzu Corp. Kyoto, Japan) having ram diameter of 42.7 mm. Circular mold was used in the present study. The processing temperature (initial temperature) and pressure were the main parameters affecting the graphite mixed, polymer composite pellets. The composites were to have good conductivity and hardness. After the processing temperature and pressure were optimized, all the samples were prepared at the processing temperature of 120°C at 70 MPa and baked at 150°C at 35 MPa for 15 min to achieve conductivity and hardness.

After baking the samples, all the samples were cooled down to room temperature at atmospheric pressure. This step helps in improving the electrical properties, minimizing after-shrinkage and enhancing quality and appearance by eliminating porosity, bubbles or blisters. Disk shaped specimens of 1.01 cm in diameter were obtained. Specimens were taken out from the mold and the surfaces of the specimens were polished by sandpaper to remove the polymer rich surface layer and to eliminate surface irregularities.

Characterization

Scanning electron microscopy

Scanning electron microscopy (SEM) of the PP-Gr composites was performed using a Hitachi scanning electron microscope (Model No. S-4700). All the samples were gold sputtered before measurement.

Hardness (Shore-D)

Durometer (model No. SRHT-501D), conforming to DIN-53505, ASTM-1706-61 and D 676-59T specifications, was used to determine the hardness of the samples.

Measurements

For the measurement of conductivity, both opposite surfaces of the samples were coated with SEM grade conductive silver paint. Copper connecting wires were bonded to the surfaces with conductive silver paint. The resistance of the samples having resistance less than 200 M Ω was measured using a conventional digital multimeter. When the resistance exceeded 200 M Ω , a Keithley Pico ammeter was used to measure the resistance. For current-voltage characteristics, Keithley 2400 source meter was used.

For studying dielectric properties, the capacitance and dissipation factor of the samples were measured in two frequency ranges, 20 Hz–5 MHz and 1 MHz–

3 GHz, using a Hewlett-Packard (HP) Impedance Analyzer (Model No. 4294) and RF Impedance/Material Analyzer (Model No. E-4991 A).

RESULTS AND DISCUSSION

Morphology

Figure 1 (a) shows the SEM image of a pure PP sample after compression molding. This image corresponds to the formation of solid PP. SEM image^{33,34} of graphite particles has confirmed their flake like shape. Figure 1(b–d) show the SEM images of composites below ϕ_c , at ϕ_c and above ϕ_c having graphite volume fraction $\phi = 0.0054$ (1 wt %), $\phi = 0.026$ (5 wt %) and $\phi = 0.052$ (10 wt %), respectively. Note that the SEM images [Fig. 1(a–d)] were taken on fractured surfaces, which were obtained by dipping the samples in liquid nitrogen and then hammering them. For $\phi < \phi_c$ in Figure 1(c), the graphite particles are far apart from each other. At $\phi = \phi_c$ in Figure 1(d), this stage clearly shows the dispersion of graphite flakes in the interfacial regions between PP particles. For $\phi > \phi_c$ the SEM image [Fig. 1(d)] shows the increasing percentage of graphite flakes in the interfacial region of PP particles.

Conductivity analysis

Figure 2(a) shows the electrical conductivity of PP-Gr composites as a function of volume fraction (ϕ) of Gr. The conductivity of the composites increases sharply near $\phi = 0.0264$. The conductivity measured clearly exhibits the insulator-conductor transition at $\phi = 0.0264$. According to percolation theory, the conductivity of composites near the insulator-conductor transition can be represented by the power law model of percolation theory^{22,23}:

$$\sigma \propto (\phi - \phi_c)^t \quad (1)$$

where σ is the conductivity of the composite, ϕ_c is the critical filler volume fraction at which percolation takes place, and t is the critical exponent of conductivity. The experimental values of conductivity are fitted to eq. (1) and log-log plot of power law is shown in Figure 2(b). The best fit of the conductivity data to the log-log plots of the power law gives $\phi_c = 0.0257$ and $t = 1.98$. The value of t for our composite system is nearly equal to 2, which is universal value of three dimensional systems.^{35–37}

I-V characteristics

I-V characteristics can be expressed as³⁸

$$I = C * V^n \quad (2)$$

where C is a constant and n is the slope in the log I-log V plot. In eq. (2), Ohm's law is satisfied by the system

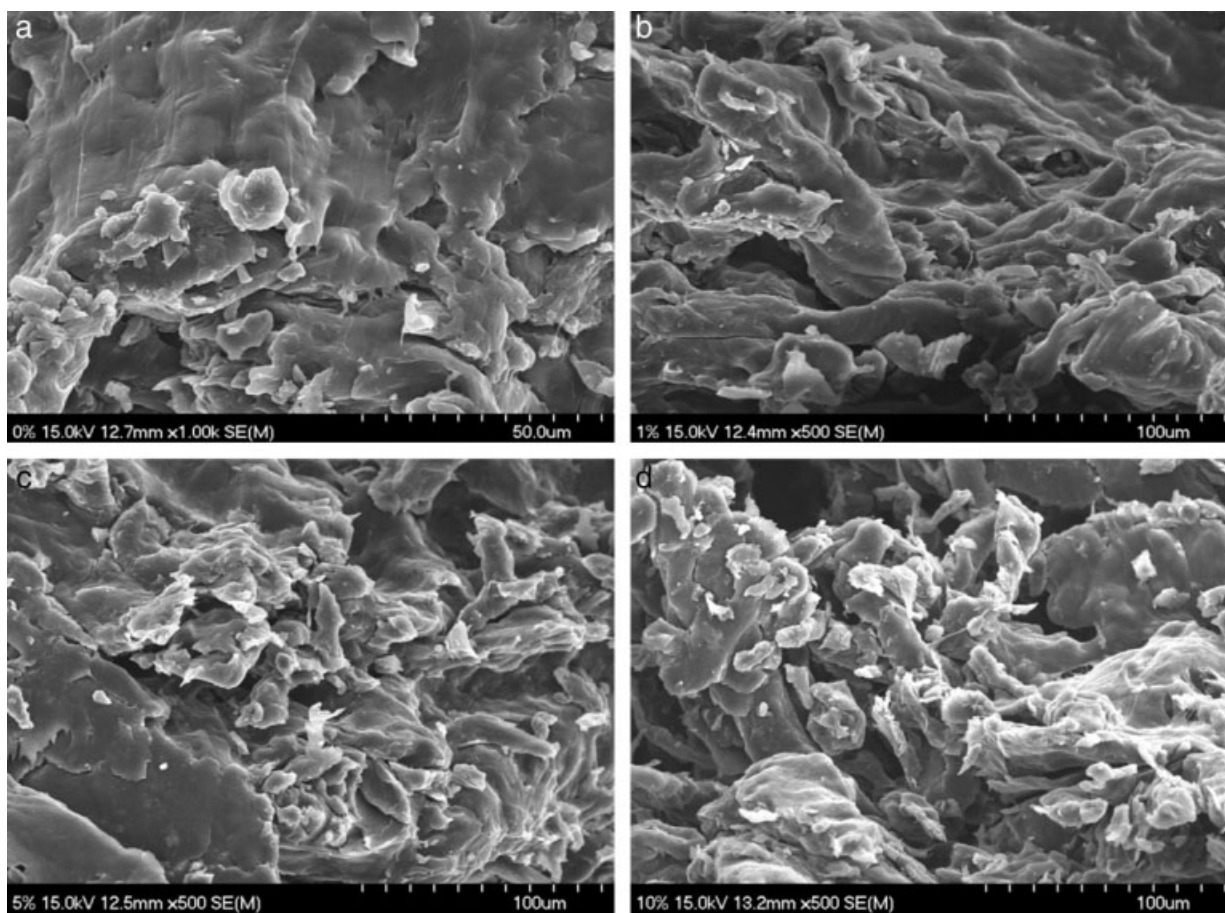


Figure 1 Scanning electron microscopy (SEM) images of (a) Fractured surface of hot compressed mold pure PP sample (b) Fractured surface of composite with $\phi = 0.0054$ and (c) Fractured surface of composite with $\phi = 0.026$ (d) Fractured surface of composite with $\phi = 0.052$.

at $n = 1$. Figure 3 illustrates the normal I-V plots of PP-Gr composite with different ϕ . The I-V plots are non-linear for the composites with $\phi = 0.015$ and 0.026 (ϕ_c). The values of n for these composites calculated using eq. (2) are 1.58 and 1.14, respectively. The conduction mechanism of conducting polymer composites is based on electron tunneling and hopping when the interparticle distance is sufficiently small. As the graphite content increases, the I-V plots becomes more linear because of electron tunneling, and hopping increases with the increase of voltage.^{33,39,40} For composites with $\phi = 0.036, 0.075$, the I-V plot becomes linear, and value of n becomes nearly equal to 1. The conduction mechanism is now supposed to be ohmic conduction because the filler particles are very close or in direct contact with each other. The variation of C with ϕ is also shown in Figure 4. The values of C increases with ϕ in a similar trend to σ , which increases with ϕ .

Dielectric properties in low frequency region

The conducting polymer composites possess a frequency (ω) dependent, complex dielectric constant ϵ^*

(ω) = $\epsilon' - i \epsilon''$ (ω). The real part ϵ' (ω) represents the relative dielectric constant and the imaginary part ϵ'' (ω) accounts for the losses. The ratio of the imaginary to the real part (ϵ''/ϵ') is the "dissipation factor," which is represented by $\tan \delta$, where δ is called as the "loss angle" and denotes the angle between the voltage and the charging current. To investigate the dielectric behavior of PP-Gr composites in the low frequency (f_L) range, the room temperature ϵ' and $\tan \delta$ of the composite were calculated in the frequency range from 20 Hz to 1 MHz.

Figure 5(a) shows the variation of ϵ' of PP-Gr composites as a function of ϕ of Gr at 40 Hz. The dielectric constant of a pure PP sample is about 2.4. The dielectric constant increases slightly up to that of a composite with $\phi = 0.015$. The ϵ' of the composite enhances greatly near $\phi = 0.026$. Large enhancement in ϵ' of composites in the low frequency region occurs due to the Maxwell-Wagner polarization originating in the insulator-conductor interfaces. This enhancement of the dielectric constant in the neighborhood of the percolation threshold is also predicted by the power law^{41–43} as follows:

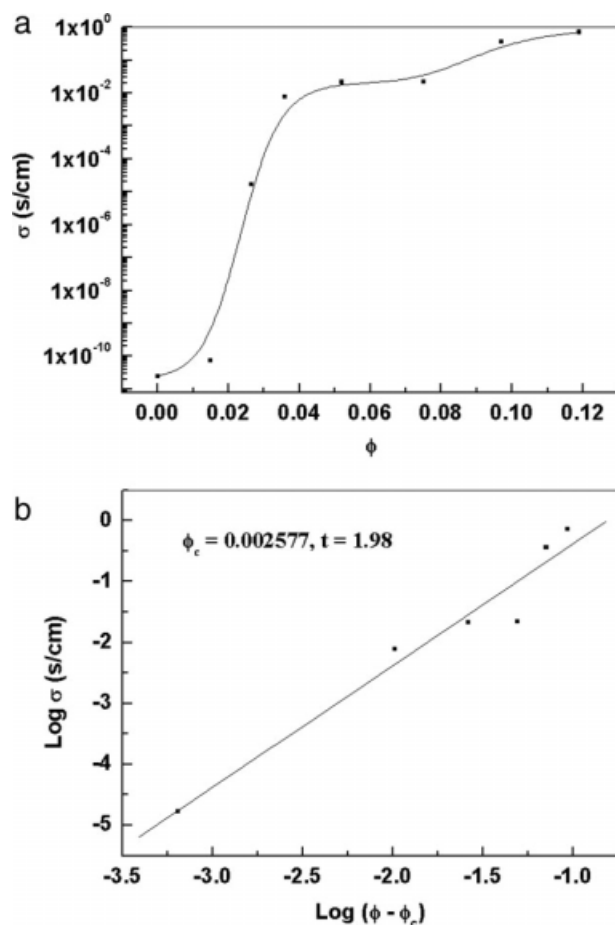


Figure 2 (a) Electrical conductivity (σ) of PP-Gr composites as a function of graphite volume fraction (ϕ). (b) Plot of $\log \sigma$ versus $\log (\phi - \phi_c)$ of PP-Gr composites.

$$\varepsilon' \propto (\phi_c - \phi)^{-s} \quad (3)$$

where s is a critical exponent. The log-log plots of eq. (3) are shown in Figure 5(b). The value of ϕ_c ($= 0.0257$) is found to be the same as that obtained for the conductivity data. The value of s is found to be

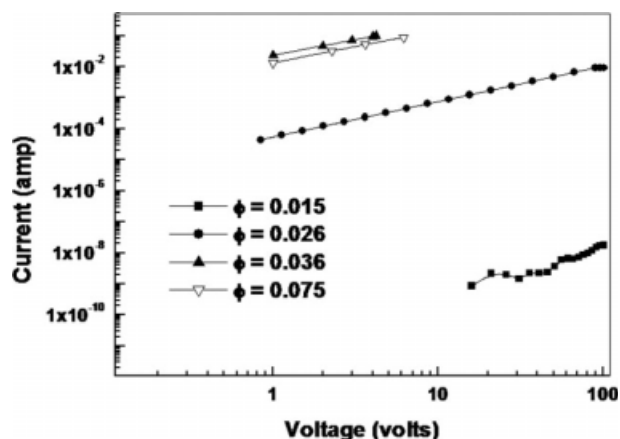


Figure 3 Plot of I-V of PP-Gr composites for different ϕ .

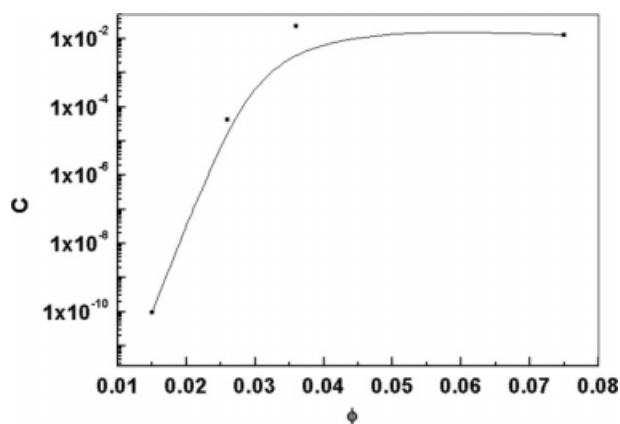


Figure 4 Variation of C of PP-Gr composites with ϕ .

1.03, which is close to the normal value given by percolation theory.⁴¹

The dependence of ε' of the PP-Gr composites on the low frequency (f_L) is shown in Figure 6(a). When $\phi < \phi_c$ for example at $\phi < 0.026$, the ε' of the composites exhibits weaker frequency dependence, but when $\phi > \phi_c$ ε' of the composites exhibits stronger frequency dependence. At low frequency, a high

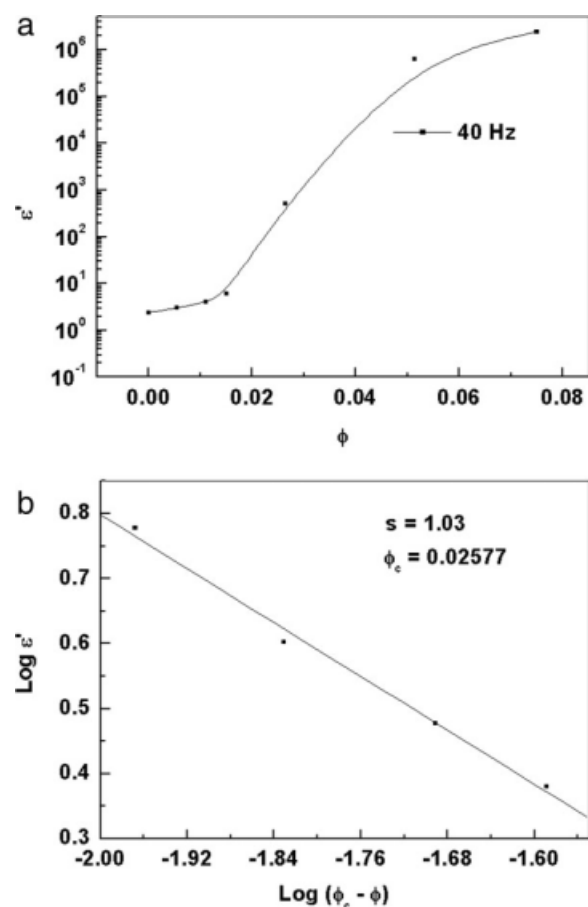


Figure 5 (a) Dielectric constant (ε') of PP-Gr composites as a function of ϕ at 40 Hz (b) Plot of $\log \varepsilon'$ versus $\log (\phi_c - \phi)$ of PP-Gr composites.

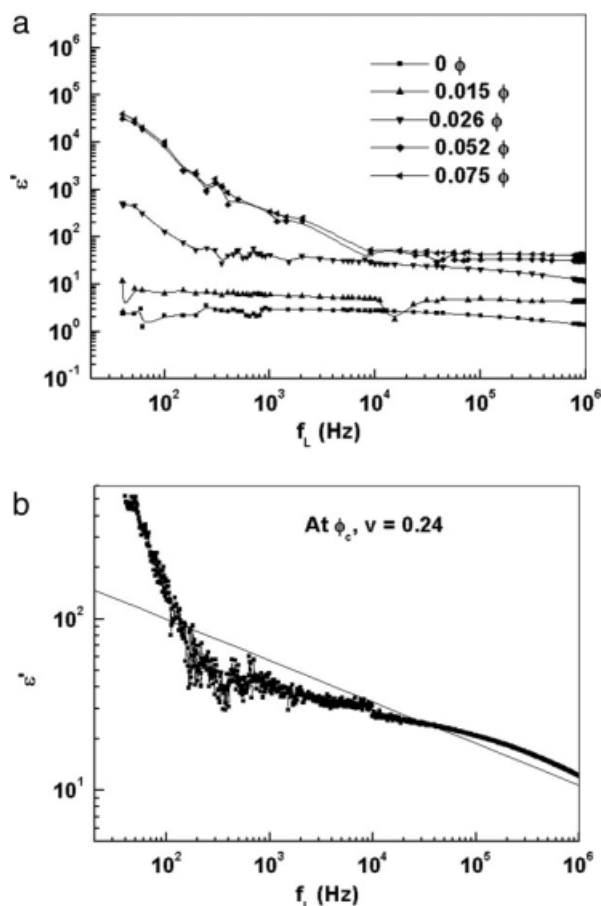


Figure 6 (a) Variation of ϵ' of the PP-Gr composites as a function of f_L for different ϕ . (b) Log-log plot of ϵ' versus f_L .

value of ϵ' of composites with $\phi \geq \phi_c$ was observed because at low frequencies, the polarization follows the change of the electric field, and the loss is minimum and the contribution the dielectric constant is maximum. The high value of $\epsilon' = 4.04 \times 10^5$ is observed at 40 Hz for the composites with $\phi = 0.075$.

High value ϵ' could be used in capacitor applications.

The frequency variation of the dielectric constant at $\phi \sim \phi_c$ as predicted by percolation theory⁴¹ is

$$\epsilon'(f, \phi_c) \propto f_L^{-v} \quad (4)$$

where v is the critical exponent. The log-log plot of eq. (4) has been shown in Figure 6(b). From the data, the value of v was found to be 0.24.

The dependence of $\tan \delta$ of the PP-Gr composites on low frequency (f_L) is shown in Figure 7. It is clear from figure that $\tan \delta$ shows complex fluctuations up to 10 KHz for composites when $\phi < \phi_c$ and after this frequency, decreases with the increase of frequency. The $\tan \delta$ of the composites with $\phi \geq \phi_c$ increases up to 3 KHz and after this frequency, the $\tan \delta$ decreases with the increase of frequency. The high value of $\tan \delta = 7.53 \times 10^4$ is observed at 600 Hz for composites with $\phi = 0.075$. High values of $\tan \delta$ of the composites could be utilized for decoupling capacitor applications.

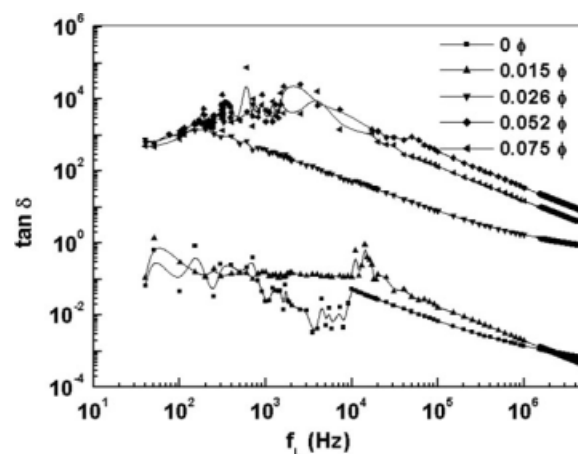


Figure 7 Variation of $\tan \delta$ of the PP-Gr composites as a function of f_L for different ϕ .

The dependence of the AC conductivity (σ_{ac}) of the PP-Gr composites on f_L is shown Figure 8(a). The AC conductivity of composites is calculated from the formula $\sigma_{ac} = \omega_L \epsilon_0 \epsilon' \tan \delta$ where $\omega_L = 2\pi f_L$. The σ_{ac} was found to increase with the increase of frequency for the composites with $\phi \leq \phi_c$ whereas remained

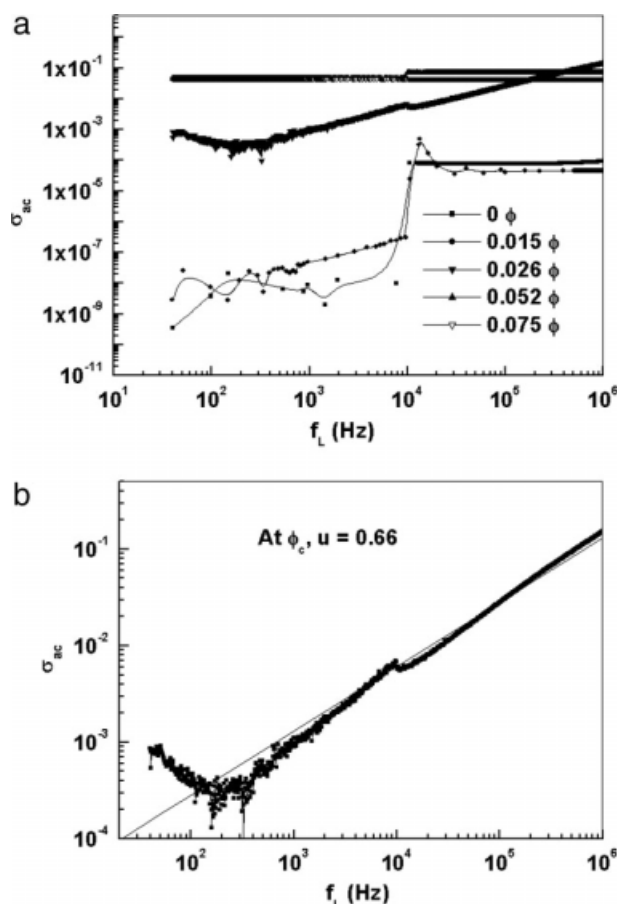


Figure 8 (a) Variation of σ_{ac} of the PP-Gr composites as a function of f_L for different ϕ . (b) Log-log plot between σ_{ac} and f_L of the PP-Gr composites.

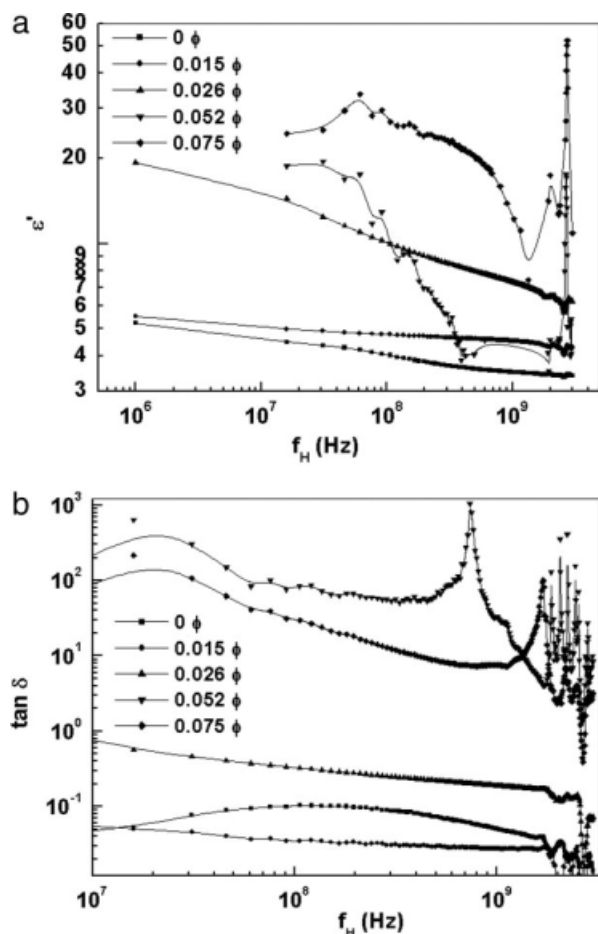


Figure 9 (a) Variation of ϵ' of the PP-Gr composites as a function of f_H for different ϕ . (b) Variation of $\tan \delta$ of the PP-Gr composites as a function of f_H for different ϕ .

constant with the increase of frequency for composites with $\phi > \phi_c$. According to the percolation theory,⁴¹ at $\phi \sim \phi_c$

$$\sigma_{ac}(f, \phi_c) \propto f_L^u \quad (5)$$

where u is the critical exponent. The log-log plot of eq. (5) is shown in Figure 8(b). From the data, the value of u was found to be $= 0.66$, which is slightly lower than the universal one ($u = 0.70$).⁴¹ The critical exponents u and v are related as $u + v = 1$. In the present case, $u = 0.66$ and $v = 0.24$ i.e., $u + v = 0.9$, which is close to the normal value given by percolation theory. The above analysis clearly confirms the percolation phenomenon in PP/Gr composites.

Dielectric properties in the high frequency region

To investigate the dielectric behavior at high frequencies (f_H) for PP-Gr composites, the room temperature ϵ' and $\tan \delta$ of the composite were calculated in the frequency range from 1 MHz to 3 GHz (radio frequency). Figure 9(a,b) show the ϵ' and $\tan \delta$ of the composite as a function of f_H for various contents of

graphite. A small variation in the ϵ' and $\tan \delta$ with respect to f_H was observed for composites with $\phi < \phi_c$ and a large variation was observed for composites with $\phi \geq \phi_c$. The ϵ' and $\tan \delta$ show a complex fluctuation with the measured f_H for composites with $\phi = 0.052$ and 0.075 . The maximum values of $\epsilon' = 52$ and $\tan \delta = 101.13$ were obtained at 2.71 GHz and 1.69 GHz, respectively, for composites with $\phi = 0.075$. High values of ϵ' and $\tan \delta$ in the radio frequency range are useful for EMI SE applications.

Figure 10 shows the σ_{ac} of the composites as a function of f_H (radio frequency) for various contents of graphite. A sharp increase in σ_{ac} with the increase of f_H was obtained for composites with $\phi = 0, 0.015$, and 0.026 . For composites with $\phi = 0.052$ and 0.075 , the σ_{ac} is high and remains constant with the increase of the frequency. So, the high AC conductivity and weak frequency dependence of the PP-Gr composites make these composites excellent anti-static media and shielding for electromagnetic or radio frequency interference of electronic devices.

EMI shielding effectiveness

An electromagnetic shield is a conductive material which attenuates (through reflection and absorption) electromagnetic energy. The electromagnetic interference (EMI) shielding effectiveness (SE) is defined as the ratio of the incident to the transmitted power and is usually expressed in decibels (dB) as follows:

$$SE = 10 \log \left(\frac{P_i}{P_t} \right) (\text{dB}) \quad (6)$$

where P_i is the incident power, and P_t is the transmitted power.

The total EMI shielding effectiveness of a sample of thickness t is given as the sum of the reflection (SE_R) and absorption (SE_A), such as $SE = SE_R +$

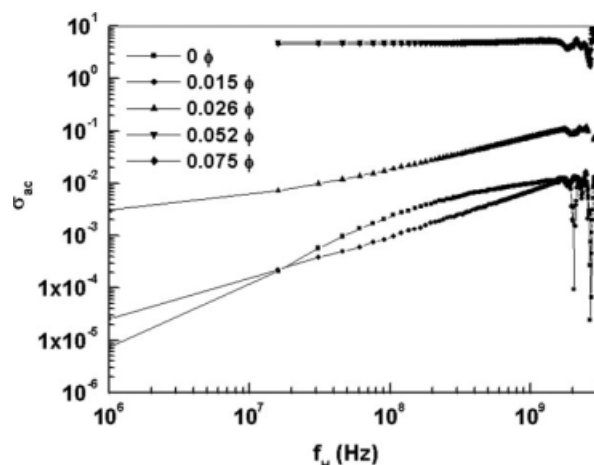


Figure 10 Variation of σ_{ac} of the PP-Gr composites as a function of f_H for different ϕ .

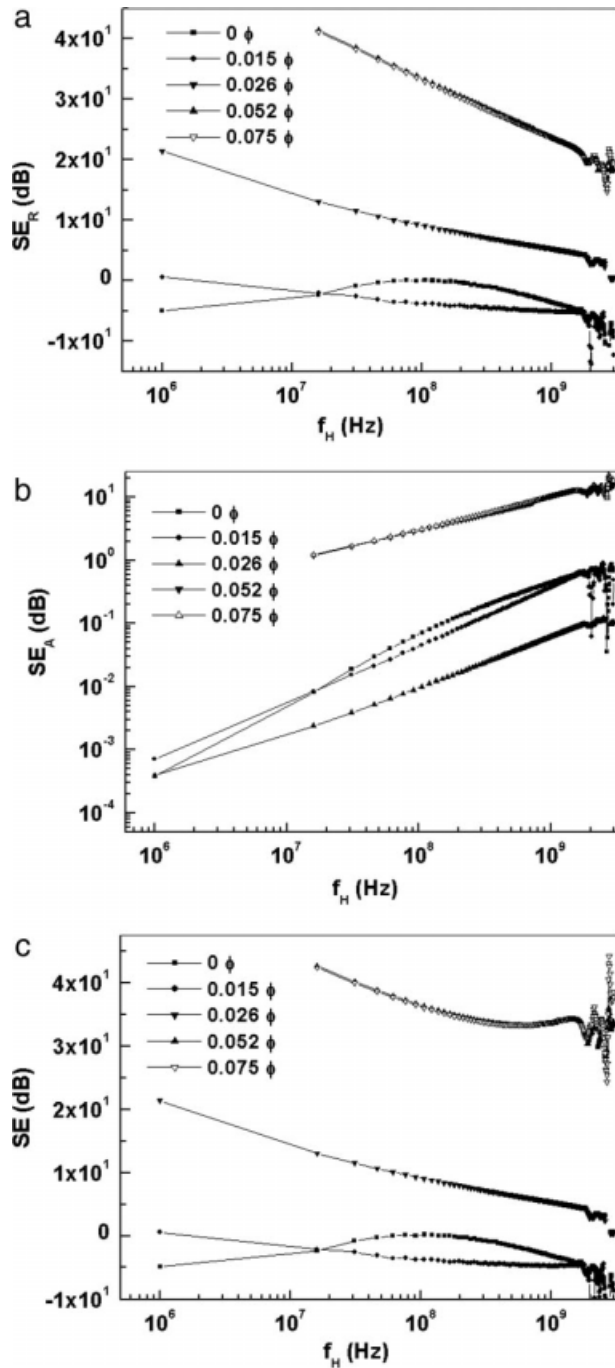


Figure 11 (a) Variation of SE_R of the PP-Gr composites as a function of f_H for different ϕ . (b) Variation of SE_A of the PP-Gr composites as a function of f_H for different ϕ . (c) Variation of SE of the PP-Gr composites as a function of f_H for different ϕ .

SE_A . According to theory,^{44,45} the reflection part of SE is

$$SE_R = 20 \log \left[\frac{\left(\frac{\sigma_{ac}}{\omega_H \epsilon_0 \mu_r} \right)^{1/2}}{4} \right] \text{ (dB)} \quad (7)$$

and the absorption part of SE is

$$SE_A = 20 \log \left[\exp \left(\frac{t}{\delta} \right) \right] \text{ (dB)} \quad (8)$$

The skin depth, $\delta = (2/\omega_H \mu \sigma_{ac})^{1/2}$, is defined as the depth to which the radiation penetrates into the material, while its intensity decreases to e^{-1} of its original strength. When calculating the shielding effectiveness SE, the magnetic permeability μ of graphite filled polymer composites, as a nonmagnetic material, can be taken as the magnetic permeability of free space, $\mu_0 = 4\pi \times 10^{-9} \text{ H cm}^{-1}$, and the relative permeability of the shielding material $\mu_r = 1$. $\omega_H = 2\pi f_H$, where f_H is the radiation frequency in MHz.

Using eqs. (7) and (8), the values of SE_R and SE_A for the PP-Gr composites were calculated. The SE_R and SE_A of the composites as a function of f_H (1 MHz–3 GHz) are shown in Figure 11(a) and 11(b), respectively. The SE_R of the composites decreases with the increase of f_H and increases with the increase of ϕ . For composites with $\phi = 0$ and 0.015, SE_R was found to be negative because of the low value of σ_{ac} of these composites because from eq. (7), SE_R depends on σ_{ac} . High value ($\sim 41 \text{ dB}$) of SE_R was obtained at 15.9 MHz for the composites with $\phi = 0.052$ because the composite have a high value of σ_{ac} . The SE_A of composites increases with f_H because SE_A is directly proportional to f_H as observed by eq. (8). The maximum value (22.37 dB) of SE_A was obtained at 2.76 GHz for composites with $\phi = 0.075$. The total SE of the composites was calculated by adding SE_R and SE_A . The SE of the composites as a function of f_H is shown in Figure 11(c). The SE of composites was found to increase with the increase of ϕ and decrease with the increase of f_H . As expected from the above data on SE_R and SE_A , the trend of SE shows that SE_R comprises a major portion of EMI SE. The maximum value (44.12 dB) of SE was obtained at 2.76 GHz for the composites with $\phi = 0.075$.

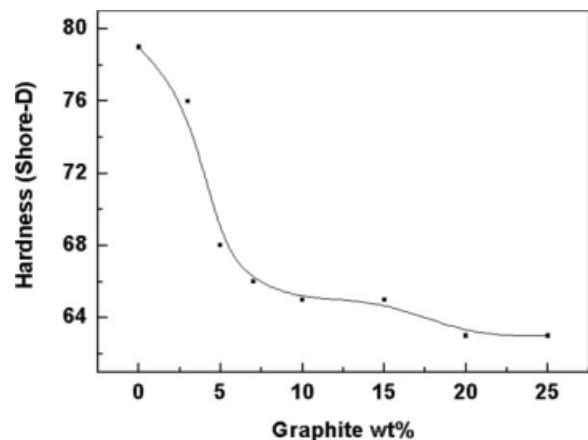


Figure 12 Variation of Shore-D hardness of PP-Gr composites as a function of Gr wt %.

Hardness

The variation of Shore-D hardness of PP-Gr composites as a function of Gr wt % is shown in Figure 12. The hardness decreases sharply with the increase in Gr content up to the percolation threshold. However, beyond threshold content, the hardness decreases slowly, possibly due to the partial prevention of fusion of polymer particles in the presence of graphite.

CONCLUSIONS

Both electrical conductivity and dielectric constant of PP-Gr composites followed the power law model of percolation theory at the percolation threshold $\phi_c = 0.0257$. At $\phi = \phi_c$, the dielectric constant and ac conductivity of PP-Gr composites at low frequencies followed the power law model of percolation theory. The dielectric constant of PP-Gr composites with $\phi \geq \phi_c$ was strongly frequency (low and high) dependent. The highest dielectric constant (4.04×10^5) was obtained at 40 Hz for composites with $\phi = 0.075$. These composites can be used in charge storing devices. The highest dissipation factor (7.53×10^4) was obtained at 600 Hz for composites with $\phi = 0.075$ and this property of the composites makes it suitable for decoupling capacitor applications. Above $\phi > \phi_c$, the composites possessed high values of AC conductivity and exhibited weak frequency (low and high) dependence, so these composites can be utilized as antistatic media and in EMI shielding applications. The main contribution of these composites to effective EMI shielding was due to reflection. The maximum value (44.12 dB) of SE was obtained at 2.76 GHz for the composites with $\phi = 0.075$. Finally, the proposed PP-Gr composites exhibited a high dielectric constant and dissipation factor with the addition of graphite content in the low and radio frequency ranges, so these composites can be utilized in charge storing devices, decoupling capacitors and electromagnetic interference (EMI) shielding applications.

References

1. Celzard, A.; Mcrae, E.; Furdin, G.; Mareche, J. F. *J Phys Codens Mater* 1997, 9, 2225.
2. Celzard, A.; Mcrae, E.; Mareche, J. F.; Furdin, G.; Sundqvist, B. *J Appl Phys* 1998, 83, 1410.
3. Zheng, Q.; Song, Y.; Wu, G.; Yi, X. *J Polym Sci Part B: Polym Phys* 2001, 39, 2833.
4. Krupa, I.; Novak, I.; Chodak, I. *Synth Met* 2004, 145, 245.
5. Lee, G. J.; Suh, K. D.; Im, S. S. *Polym Eng Sci* 1998, 38, 471.
6. Chodak, I.; Krupa, I. *J Mater Sci Lett* 1999, 18, 1457.
7. Yang, G.; Tang, R.; Xiao, P. *Polym Compos* 1997, 18, 477.
8. Nakamura, S.; Saito, K.; Sawa, G.; Kitagawa, K. *Jpn Appl Phys* 1997, 36, 5163.
9. Li, Y. J.; Xu, M.; Feng, J. Q.; Dang, Z. M. *App Phys Lett* 2006, 89, 72902.
10. Bloor, D.; Donnelly, K.; Hands, P. J.; Laughlin, P.; Lussey, D. *J Phy D Appl Phy* 2005, 38, 2851.
11. Ramasubramaniam, R.; Chen, J. *Appl Phys Lett* 2003, 83, 2928.
12. Kymakis, E.; Amaratunga, G. A. J. *J App Phys* 2006, 99, 84302.
13. Kilbride, B. E.; Coleman, J. N.; Fraysse, J.; Fournet P., Cadek M., Drury A., Hutzler, S. *J Appl Phys* 2002, 92, 4024.
14. Calame, J. P. *J Appl Phys* 2006, 99, 084101.
15. Rao, Y.; Wong, C. P. *J App Polym Sci* 2004, 92, 2228.
16. Heaney, M. B. *Appl Phys Lett* 1996, 69, 2602.
17. Strumpler, R. *J Appl Phys* 1996, 80, 6091.
18. Chen, Q.; Du, P.; Lu, J.; Weng, W.; Han, G. *App Phys Lett* 2007, 91, 22912.
19. Lu, J.; Moon, K. S.; Xu, J.; Wong, C. P. *J Mater Chem* 2006, 16, 1543.
20. Chung, D. D. L. *Carbon* 2001, 39, 279.
21. Wu, J.; Chung, D. D. L. *Carbon* 2002, 40, 445.
22. Stauffer, D.; Aharony, A. *Introduction to Percolation Theory*; Taylor and Francis: London, 1992, 89.
23. Efros, A. L.; Shklovskii, B. I. *Phys. Stat Sol* 1976, 76, 475.
24. Shen, J. W.; Chen, X. M.; Huang, W. Y. *J Appl Polym Sci* 2003, 88, 1864.
25. Kalaitzidou K.; Fukushima, H.; Drzal, L. T. *Compos Sci Techn* 2007, 67, 2045.
26. Zois, H.; Apekis, L.; Omastova, M. 10th International Symposium on Electrets 1999, 529.
27. Dweiri, R.; Sahari, J. *J Power Sources* 2007, 171, 424.
28. Bai, Y.; Cheng, Z. Y.; Bharti, V.; Xu, H. S.; Zhang, Q. M. *App Phys Lett* 2000, 76, 3804.
29. Dang, Z. M.; Fan, L. Z.; Shen, Y.; Nan, C. W. *Mater Sci Eng B* 2003, 103, 140.
30. Choi, H. W.; Heo, Y. W.; Lee, J. H.; Kim, J. J.; Lee, H. Y.; Park, E. T.; Chung, Y. K. *Appl Phys Lett* 2006, 89, 132910.
31. Huang, C.; Zhang, Q. M.; Su, Ji. *Appl Phys Lett* 2003, 28, 3502.
32. Huang, C.; Zhanga, Q. M.; Debotton, G.; Bhattacharya, K. *Appl Phys Lett* 2004, 84, 4391.
33. Panwar, V.; Sachdev, V.K.; Mehra, R.M. *Eur Polym J* 2007, 43, 573–585.
34. Panwar, V.; Mehra R.M. *Polym Eng Sci*, to appear.
35. Lux, F. *J Mater Sci* 1993, 28, 285.
36. Kirkpatrick, S. *Rev Mod Phys* 1973, 45, 574.
37. Balberg, I. *Phys Rev Lett* 1983, 59, 1305.
38. Mamunya, Y. P.; Muzychenko, Y. V.; Pissis, P.; Lebedev, E. V.; Shut, M. I. *Polym Eng Sci* 2002, 42, 90.
39. Feng, J.; Chang, C. M. *Polym Eng Sci* 1998, 38, 1649.
40. Sachdev, V. K.; Panwar, V.; Singh, H.; Mehra, N. C.; Mehra R. M. *Phys Stat Sol* 2006, 203, 386.
41. Nan, C. W. *Prog Mater Sci* 1993, 37, 1.
42. Chitame, C.; McLachlan, D. S. *Phys Rev B* 2003, 67, 24206.
43. Bergman, D. J. *Phys Rev Lett* 1980, 44, 1285.
44. Paligova, M. J.; Saha, V. P.; Kresaleka, V.; Stejskal, J.; Quadrat, O. *Physica A* 2004, 335, 421.
45. Colaneri, N. F.; Shacklette, L. W. *IEEE Trans Instrum Meas* 1992, 41, 291.