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Study of the electrical properties of polystyrene–foliated graphite composite

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The present paper reports the development of polystyrene–foliated graphite composites via a hot compression moulding technique. Foliated graphite, as obtained by the sonication of expanded graphite, was used as a filler. A distinct percolative behaviour was observed in the variation of conductivity of the composites as a function of graphite concentration. At the percolation concentration of graphite content (0.02 vol. fraction), the conductivity is found to be ca. 10^{-5} S/cm. The percolation behaviour was analyzed using the generalized effective media equation. An estimation of the interparticle distance between the filler particles/clusters in the composites was made. The analysis of current–voltage characteristics revealed that near the percolation threshold the electrical transport is due to tunnelling of charge carriers, while above the threshold it is ohmic in nature.

Key words: *polymer composites; electrical properties; percolation threshold; scanning electron microscopy (SEM)*

1. Introduction

The electrical insulating behaviour of most polymeric materials is well known. However, conductive fillers can be incorporated as a second phase into these matrices, leading to an increase in the conductivity of the resulting composites [1–3]. The properties of these composites are mainly varied with the filler content [4–6]. When the filler content reaches a critical value (the so-called percolation threshold), a sharp transition in the conductivity of the composites occurs with a slight increase of the filler content [7, 8]. Nanosize fillers significantly modify the properties of polymer composites and even generate certain new properties that cannot be derived from conventional fillers [9]. The incorporation and dispersion of nanosize conducting fillers is assumed

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to be better as compared with conventional microsize fillers and thus a low percolation threshold and higher conductivity are achieved.

Naturally abundant graphite flakes have been widely used as conducting fillers for polymer composites. Conventional graphite fillers are usually microsize powders. In order to fabricate a composite with a satisfactory conductivity, loadings of fillers are usually as high as 20 wt. % or even higher. This often results in poor mechanical properties of the material. Polymer composites made from graphite nanosize powder or nanosheets may possess promising properties, especially good electrical conductivity at low filler loading, corresponding to low percolation concentration of the filler. Various theories/models have been proposed to interpret the electrical conductivity behaviour of polymer composites [3, 8, 10].

The percolation threshold concentration of filler depends on the dispersion of the conducting filler into the polymer matrix and the preparation technique of such composites. The conducting polymer composite can be prepared using either hot compression moulding or in-situ polymerization. In hot compression moulding [11–13], the filler particles are pre-localized [14, 15] onto the polymer particles before compression. In the case of in-situ polymerization, the polymer particles are dispersed into the monomer before polymerization. The monomer is gradually polymerized with the addition of an initiator, along with proper sonication of filler particles to achieve better dispersion. The effect of filler content on the conductivity of the composite is generally analyzed by using percolation theory for both types of composites. In the hot compression mouding technique, the conductivity of the composites depends on various parameters of the individual polymer and graphite particles. The main parameters which are primarily responsible for the electrical behaviour of the composites are (i) conductivity, (ii) size, (iii) structure and (iv) orientation. For such a system, McLachlan [16, 17] has given a generalized effective media (GEM) equation. The conduction mechanism of various polymers/graphite nanosheets fabricated using in-situ polymerization process has been reported earlier by several groups [18, 19]. The schematic representation of the dispersion of nanosize filler has been reported by Chen et al. [20] using a poly(methyl methacrylate)–exfoliated graphite composite system.

This paper deals with the preparation of polystyrene–foliated graphite composites by the hot compression moulding. Foliated graphite (FG) was used as filler. The foliated graphite was characterized using scanning electron microscopy (SEM) and X-ray diffraction (XRD). Variation of room temperature conductivity of the composite with filler content was investigated. The percolation concentration was estimated using a power law model of conductivity. The electrical conductivity versus filler volume fraction data was also analyzed using the GEM equation. The interparticle distance between the filler particles/clusters using the sizes of the insulating and conducting components of the system was calculated. These results were ascertained by SEM morphology of fractured composites. The effect of filler content on the current–voltage (I–V) characteristics was also investigated. The mechanical strength of the composites was estimated in terms of the D-shore hardness.

2. Experimental

Materials. The matrix polymer used in the present study is a commercial grade polystyrene (PS) manufactured by the Reliance Industry Ltd., Bangalore, India, supplied in the form of granules. The PS granules were ground to obtain finer particles using an Aurther H. Thomas type Willey grinder. The scanning electron micrographs (SEM) of PS particles and foliated graphite coated PS particles are shown in Fig. 1. It is seen from the images that the PS particles have an average size of 212–250 μm and the shape close to an ellipsoid. It is also seen from Fig. 1b that there is no significant change in the shape of the polymer due to tumble mixing or after pre-localization of filler. The glass transition temperature (T_g) of PS, as determined from differential scanning calorimetric measurements, was found to be 100 °C.

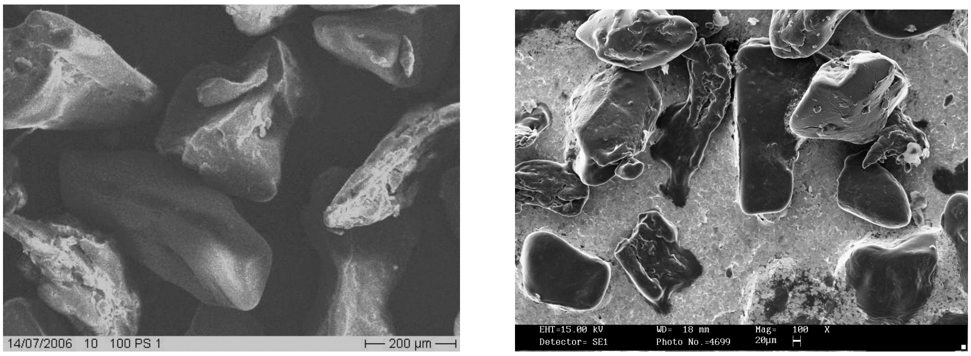

Fig. 1. SEM images of PS particles (left) and FG coated PS particles (right)

Natural graphite flakes (NFG) with average particle size of 10–20 μm, supplied by the Graphite India Ltd., were used as the source of foliated graphite. Conductivity of the graphite flakes was 1.33×10^4 S/cm with the density of 1.75 g/cm^3. FG is used as the conducting filler to prepare PS/FG composites.

Expansion of graphite. Graphite is a form of carbon where the carbon atoms are bonded in layers with van der Waals forces in between the layers. This structure allows intercalation by additional atoms or molecules which occupy spaces between the carbon layers. Graphite can be intercalated by exposure to an appropriate chemical reagent. Intercalation can be performed by immersing NFG in the mixture of concentrated HNO_3 and H_2SO_4. The reaction is as follows [21]:

$$n(\text{graphite}) + n(\text{H}_2\text{SO}_4) + 0.5n(\text{O}) \rightarrow n(\text{graphite}\cdot\text{HSO}_4) + 0.5n\text{H}_2\text{O} \qquad (1)$$

where (O) is the oxidant and (graphite·H_2SO_4) is the graphite intercalated compound (GIC). The resulting GIC comprises carbon layers and intercalated layers stacked on top of one another in a periodic fashion. The number of carbon layers between each pair of intercalated layers is the stage number. To obtain GIC from NFG, concentrated sulfuric and nitric acid (4:1, v/v) were mixed with NFG at room temperature and stirred continuously for 16 h. Acid-treated natural graphite was then washed with water and dried at 100 °C to obtain GIC. The expansion of GIC can be achieved by using a thermal shock [22]. In the present case, GIC was exposed in a microwave oven for 20 s to obtain expanded graphite (EG). The SEM image of EG is shown in Fig. 2 at higher magnification (×4000). The inset in Fig. 2 shows the SEM image at lower magnification (×15). It is seen from the inset that the GIC has expanded about hundred times along the *c*-axis. The EG is a loose and porous vermicular product of greatly decreased density. The structure of EG basically comprised parallel boards which were completely torn into sheets 80–150 nm thick.

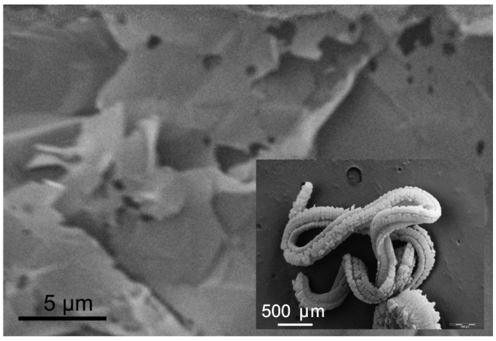

Fig. 2. SEM images of expanded graphite. The magnification of the main photograph is 4000×, that of the inset 15×

Structure of foliated graphite. Foliated graphite was obtained by sonication of 70% aqueous–alcohol solution of EG in an ultrasonic bath for 8 h [23, 24]. The resulting dispersion was then filtered and dried to get FG. Figure 3 shows a SEM image of FG at magnification of 5000×. It is seen from the image that the foliated graphite has a sheet type structure. The range of thickness and diameter of FG is estimated to be 80–150 nm and 5–10 μm, respectively. The sheet structure of foliated graphite, shown in Fig. 4, was also observed by the transmission electron microscopy (TEM) in a normal mode. The selected area diffraction (SAD) of the FG (the inset of Fig. 4) indicates the polycrystalline nature of foliated graphite.

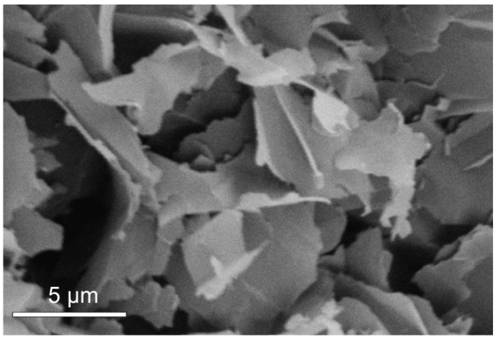

Fig. 3. SEM image of foliated graphite

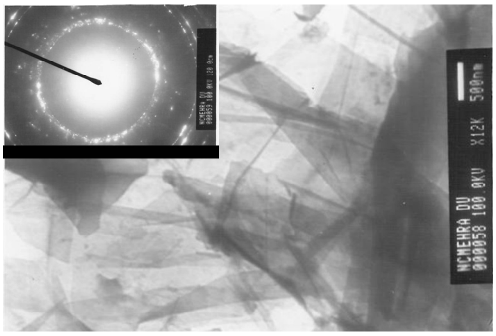

Fig. 4. TEM images of foliated graphite in normal mode and in diffraction mode (inset)

Preparation of composites. Initially, the as obtained foliated graphite was mixed with the polymer by pastel mortar. The mixture was further tumble mixed [13] thoroughly for 4 h at room temperature. This process involves coating of conducting foliated graphite on the surface of PS particles, referred as pre-localization of the conductive phase. Prolonged mixing improves the homogeneity of the spatial distribution of the conductive particles. The localized foliated graphite onto the PS particles was compacted by hot compression moulding in a piston cylinder assembly using a hydraulic press (Shimadzu Corp. Kyoto, Japan) having the ram diameter of 42.7 mm. The compression moulding operation was performed first at 90 °C for 15 min under 105 MPa (curing), then at 120 °C for 15 min under 20 MPa (baking). A series of disk shaped specimens of PS/FG conductive composites with varying filler contents from 0 to 0.115 volume fraction of FG were prepared. The density of the compression moulded pure PS and pure FG pellet was found to be 1.02 g/cm^3 and 1.97 g/cm^3, respectively. The sample diameters were 1.004 cm while thicknesses were ca. 0.3 cm for electrical measurements and ca. 0.5 cm for accurate and reliable hardness measurements.

Characterization and measurements. Both surfaces of the pellets were polished with sandpaper to remove a graphite rich surface layer and to eliminate surface irregularities. The conducting silver paint 'conductrox 3347 AG conductor' coatings supplied by the Ferroelectric Materials, USA, were used as the electrical contact. Pellets were sealed in air free polyethylene bags prior to testing in order to avoid atmospheric and humidity effects. A digital multimeter was used for samples having electrical resistance lower than 200 MΩ. However, for the samples having resistance higher than 200 MΩ, a Keithley Pico ammeter (Model DPA III) was used. Current-voltage measurements were performed with a Keithley 2400 source meter. The JEOL JSM 840 SEM was used for the analysis of morphology of the foliated graphite and composite polymer. X-ray diffraction measurements were done using CuK$_\alpha$ radiation ($\lambda = 0.154$ nm) operated at 40 kV and 40 mA in the 2θ range of 20–80°. A Scientico Hardness tester (Durometer, model No. SRHT-501D), conforming to ASTM 1706-61 and D 676-59T specifications, was used to determine the hardness of the samples.

3. Theory

The most popular theory used to explain the electrical conductivity of polymer composites is the percolation theory, based on the power law model [25, 26].

$$\sigma_{pc} = \sigma_{fg}(\phi - \phi_c)^t \tag{2}$$

where σ_{pc} is the electrical conductivity of the polymer composite, ϕ is the filler content in volume fraction, ϕ_c is the percolation threshold concentration of the filler, t is the universal exponent determining the power of the conductivity increase above ϕ_c and σ_{fg} is considered to be the conductivity of the filler, i.e. foliated graphite in the present case. The value of t is found to be in the range 1.5–2.0 [25–28] which is invariant on the polymer matrix, conducting filler, magnitude of the two different components, structure of the resulting composite, etc. Various research articles with different polymer matrix and filler, such as low density polyethylene and carbon black [29], poly(vinylidene fluoride) and stainless steel fibre [30], ethylene butylacrylate and carbon black [31] and polyurethanes and single walled carbon nanotubes [32], show the range of the composite conductivity (10^{-15} – 10 S/cm) along with filler volume fraction (0–0.5). Almost entirely the same range of conductivity as well as the filler volume fraction is assumed to be responsible for the so-called universal nature of t. In some cases, a higher amount of filler content is required to achieve the above-mentioned conductivity; however, t has not been affected due to the relative value of $(\phi - \phi_c)$ with conductivity. The nature of the electrical behaviour is more or less similar beyond ϕ_c with a sharp increase of 6–8 orders in conductivity against filler content. No further information has been disclosed from the power law model.

The variation of conductivity of the composite as a function of filler content can be critically examined by the GEM equation since it takes into account the intrinsic conductivities, geometries and arrangement of the filler and polymer particles along with their orientations to an applied electric field. Prior to applying the GEM equation, it is assumed that (i) the binary system is homogeneous, (ii) the particles are in contact with each other (no voids) and (iii) the electrical contact potential between insulating and conducting particles is negligible. McLachlan postulated a generalized effective medium (GEM) equation [16, 17] for the electrical conductivity of the binary composite polymer system. The GEM equation is written as

$$\frac{f\left(\sigma_{ps}^{t}-\sigma_{pc}^{t}\right)}{\sigma_{ps}^{t}+A\sigma_{pc}^{t}}+\frac{\phi\left(\sigma_{fg}^{t}-\sigma_{pc}^{t}\right)}{\sigma_{fg}^{t}+A\sigma_{pc}^{t}}=0 \tag{3}$$

where σ_{ps}, σ_{pc} and σ_{fg} are the conductivities of the polymer composite, the polymer matrix (polystyrene) and the conducting filler (foliated graphite) respectively, f and ϕ are the volume fractions of the polymer and conducting filler, thus $f + \phi = 1$. A is defined as

$$A=\frac{\left(1-\phi_c\right)}{\phi_c}=\frac{f_c}{\left(1-f_c\right)} \tag{4}$$

In the extreme limiting condition, the low component conductivity tends to zero, causing Eq. (3) to reduce to

$$\frac{\sigma_{pc}}{\sigma_{fg}}=\left(1-\frac{f}{f_c}\right)^{t} \tag{5}$$

For the critical filler volume concentration $\phi_c << 1$, Eq. (5) reduces to Eq. (2).

The critical filler concentration is related to the geometries and orientations of both the components [16];

- for an oriented ellipsoid

$$\phi_c = \frac{L_\phi}{(1 - L_f + L_\phi)} \tag{6}$$

- for a random ellipsoid

$$\phi_c = \frac{m_f}{(m_f + m_\phi)} \tag{7}$$

where L_ϕ and L_f are the demagnetization constants of foliated graphite and polystyrene particles, respectively. L stands for the combination of particle shape and orientation to the applied field. The value of L is 1/3 for spherical particles; 1 and 0 for the fiber/layer-shaped component oriented along and perpendicular to the compaction direction, respectively. m_ϕ and m_f are parameters for a random case. Exponent t in Eq. (3) is also related to oriented and random ellipsoids, respectively, by the following equations [16]

$$t = \frac{1}{1 - L_f + L_\phi}, \qquad t = \frac{m_f m_\phi}{m_f + m_\phi} \tag{8}$$

From Eqs. (6)–(8), L and m are given as

$$L_\phi = \frac{\phi_c}{t}, \qquad L_f = 1 - \frac{1 - \phi_c}{t} \tag{9}$$

$$m_\phi = \frac{t}{\phi_c}, \qquad m_f = \frac{t}{(1 - \phi_c)} \tag{10}$$

The electrical conductivities in function of filler content for the composite under study were fitted to the GEM equation by four-parameter computer programming [33].

4. Results and discussion

4.1. X-Ray diffraction of foliated graphite

The X-ray diffraction pattern of foliated graphite is shown in Fig. 5. A strong c-axis oriented diffraction peak (002) is observed at 26.78°. The diffraction pattern confirms the polycrystalline nature of foliated graphite. The full width at half maximum (FWHM) of the peak is 0.240°. The crystallite size of the FG was also estimated from the FWHM of the (002) diffraction peak by using Scherrer's relation [34]

$$l = \frac{0.94\lambda}{\beta \cos\theta} \tag{11}$$

where λ is the wavelength of X-ray radiation, θ is the Bragg angle of the (002) peak and β is the angular width of the (002) peak at a half of its maximum intensity (FWHM). The grain size of the polycrystalline foliated graphite was found to be 34 nm.

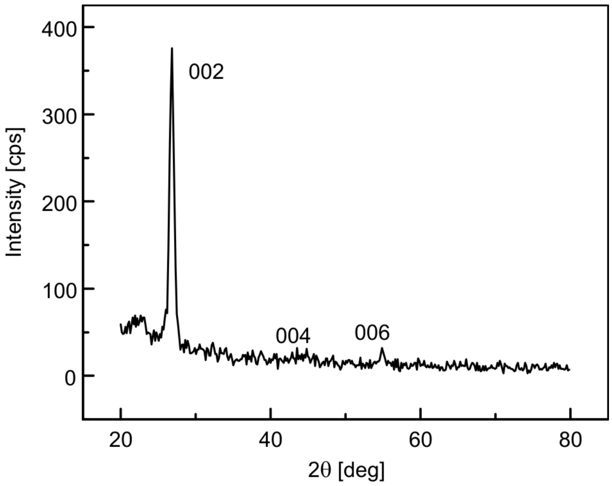

Fig. 5. X-ray diffraction pattern of foliated graphite

4.2. Electrical properties of composites

The electrical conductivity σ_{pc} of FG filled PS composites as a function of graphite volume fraction ϕ is shown in Fig. 6. It is seen from the figure that σ_{pc} increases by more than six orders of magnitude when the graphite content reaches 0.017 volume fraction (3 wt. %) of FG. The electrical conductivity reached 10^{-1} S/cm at the graphite content of 0.041 volume fraction (5 wt. %) and thereafter saturation in conductivity was observed for higher concentrations of filler.

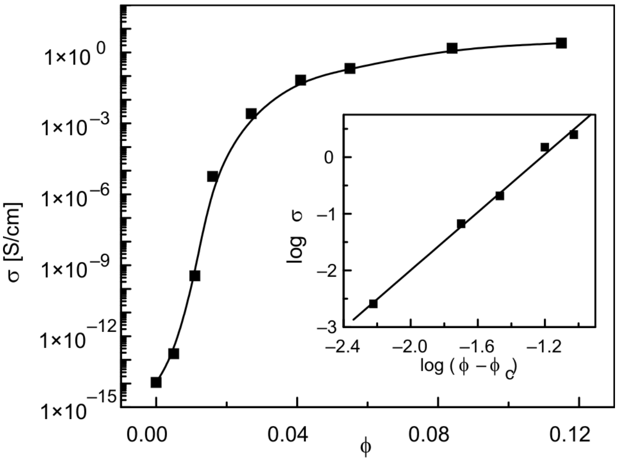

Fig. 6. Dependence of electrical conductivity on the filler content and in the inset dependence of logσ_{pc} on log ($\phi - \phi_c$) of PS/foliated graphite composites

4.3. Power law model

The conductivity data with the filler volume fraction were analyzed using the power law model (Eq. (2)). The plot of log σ_{pc} in function of log $(\phi - \phi_c)$ is shown in the inset of Fig. 6. The best linear fit was found for $\phi_c = 0.02$ volume fraction of graphite with minimum standard deviation ($\delta = 0.101\%$). The values of t and σ_{fg} as estimated from the slope and intercept of the line, are found to be 2.56 and 1.33×10^3 S/cm, respectively. The value of σ_{fg}, as obtained from the power law model, is one tenth of the conductivity of filler particles (1.33×10^4 S/cm). The observed value of $t = 2.56$ is higher than as predicted by the power law model. Probably the easy and fine dispersion of two-dimensional foliated graphite is responsible for the better segregated chains which causes a large increase in conductivity beyond the critical filler concentration, giving rise to higher values of t. Liu et al. [32] and Yuen et al. [35] have found large t values for polyurethanes and single walled carbon nanotube systems, and for epoxy and multi walled carbon nanotube systems, respectively.

4.4. GEM analysis

Using the four-parameter computer fitting program, t, ϕ_c, σ_{pc} and σ_{fg} were evaluated from the best fits and, finally, L_ϕ, L_f, m_ϕ, and m_f were calculated where L and m, as mentioned above, are the orientation based parameters. The composite conductivity, as estimated from the GEM equation with filler concentration, was further compared with the experimental conductivity in Fig. 7. Various parameters, as fitted and estimated from the GEM equation, are compared with the results obtained from the power law model in Table 1.

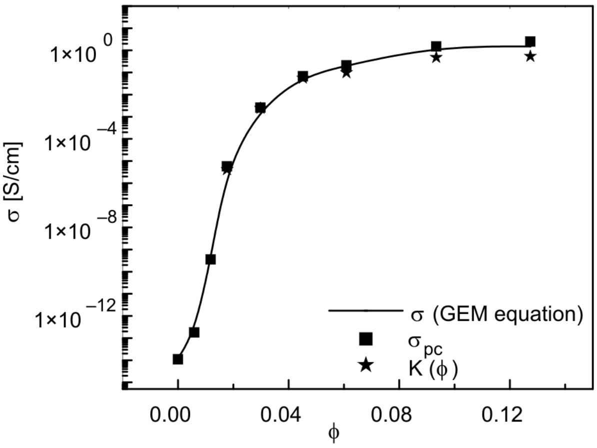

Fig. 7. Comparison of $\sigma_{pc}(\phi)$ with GEM conductivity and K of PS/foliated graphite composites

Table. 1. Parameters fitted and estimated from GEM equation compared with the power law model

Parameter	GEM equation	Power law model
t	1.83	2.56
Φ_c	0.019	0.02
σ_{ps} [S/cm]	1.354×10^{-14}	–
σ_{fg} [S/cm]	176.881	1.33×10^{3}
L_ϕ	0.01	–
m_ϕ	96.316	–
L_f	0.47	–
m_f	1.865	–
δ[%]	0.85	0.101

In the PS–foliated graphite composite, the smaller particles, i.e. FG, play the conducting role and are thus responsible for the percolation phenomenon as well as for the electrical conductivity of the system. Based on the preparation of the composite, filler particles get coated onto the matrix during dry mixing, referred as pre-localization of the filler [36], followed by compaction in a hot compression moulding, causing orientation of the filler particles during moulding, and thus forming a segregated network. A high aspect ratio between the insulating and conducting particles further ensures the segregated distribution. The value of L_ϕ as estimated from the GEM equation is very low, suggesting that almost all the filler particles are oriented perpendicular to the compaction direction, and was also confirmed from the SEM images, as shown in Fig. 8.

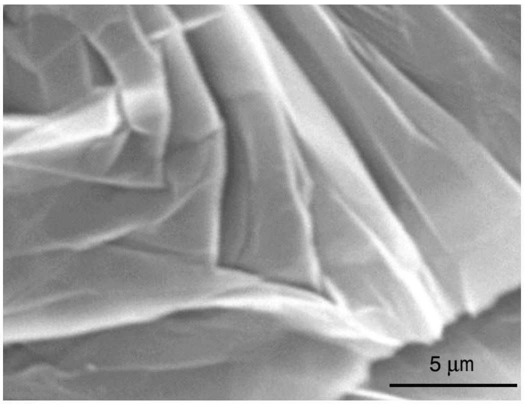

Fig. 8. SEM image of freeze fractured PS–foliated graphite composite for 0.115ϕ

Apparently, a layered type of structure of the filler particles as seen in the SEM morphology, further justifies a low value of L_ϕ. The estimated value of L_ϕ is 0.01 (close to 0), ensures the layered type of filler structure is oriented perpendicular to the compaction direction. The value of L_f is found to be 0.47. It may be mentioned that for spherical particles the GEM theory assumes L_f to be 0.333. The higher value of L_f, in the present case, as estimated from the parameters of the GEM equation, could be attributed to a non-spherical shape (close to ellipsoid) of the polymer, as seen in Fig. 1a. The percolation threshold concentration of the filler is 0.019 volume fraction of FG from the four-parameter fittings of the GEM equation with the minimum standard deviation ($\delta = 0.85\%$). The value of ϕ_c obtained from both the power law model and GEM equation is almost the same as the content of filler and is close to the experimental volume fraction (0.017, 3 wt. %).

It is seen from Table 1 that there is a considerable difference in the values of t as obtained from the GEM equation and power law model. However, the value of t obtained from the GEM equation is close to the universal value of the exponent.

4.5. Interparticle distance

The motivation behind the concept of a percolation threshold is the creation of a connected network of filler particles in between the polymer matrix by forming a direct contact or very small interparticle distance between the filler particles. The interparticle distance (d_{fg}) is estimated using the expression [37]

$$d_{fg} = r_{fg}\left(3.95\left(\frac{r_{ps}}{r_{fg}}\right)^{-1/2} f^{1/6}\phi^{-1/2} - 2\right) \tag{12}$$

where r_{ps} and r_{fg} are the average radii of polymer and filler particles, respectively. The above equation has been obtained under the assumption of spherical polymer and filler particles. In the present case, polymer particles can be assumed to be spherical; however, the filler particles (graphite) are in the form of sheets. The average sheet thickness (0.115 μm) is the value used for r_{fg}. Using the average radius of the polymer (ca. 115 μm), the interparticle distance was found to be negative for $\phi \geq \phi_c$. A negative interparticle distance represents the direct contact of the filler particles with each other. Below the percolation concentration of the filler, the interparticle distance becomes positive.

The cross-sectional SEM image (magnification 5000×) of the freeze-fractured PS–foliated graphite composite for $\phi > \phi_c$ (volume fraction of FG 0.115) is shown in Fig. 8. The image clearly shows the layers of connected FG over the polymer matrix. The higher particle size ratio of the PS resin and foliated graphite ensures segregated distribution of filler onto the polymer matrix pertaining to a much lower percolation threshold concentration. Below ϕ_c, the filler particles are far apart with no connectivity amongst each other giving rise to very low conductivity.

4.6. Current–voltage characteristics

Current–voltage (I–V) characteristics can be expressed as [38]

$$I(\phi, V) = K(\phi)V^n \tag{13}$$

where $K(\phi)$ represents the conductivity of the composite at a given value of ϕ, and n is the slope in the $\log I$–$\log V$ plot. In Equation (13), Ohm's law is fulfilled in the system at $n = 1$.

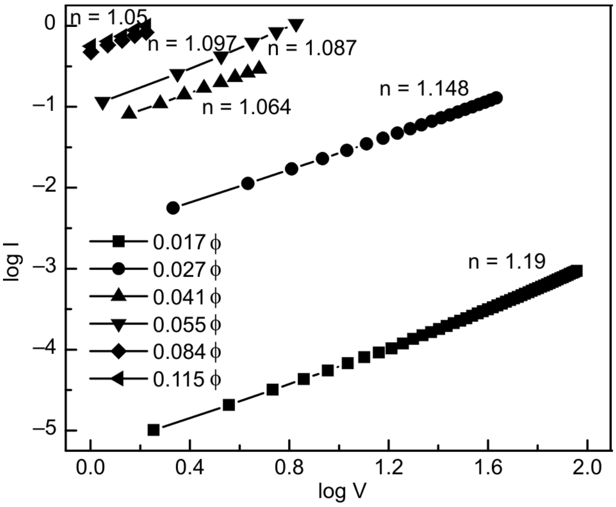

Fig. 9. $\log I(V)$ of PS/foliated graphite composites

Figure 9 illustrates the $\log I$–$\log V$ plots of PS–foliated graphite composite with different compositions. It is seen from the figure that the value of $n = 1.23$ at ϕ_c decreased to 1 at $\phi \approx 0.041$. The transition of the conduction mechanism from non-linear to ohmic is generally understood in terms of the distance and the number of tunnelling gaps in the percolating network [39]. At a sufficiently higher filler concentration for $\phi >> \phi_c$, the tunnelling gaps start to change into ohmic contacts. This effect is clearly visible in the SEM image of the polymer composite for $\phi = 0.115$ (Fig. 8). The variation of $K(\phi)$ and σ_{pc} with ϕ is also shown in Fig. 7. It is seen from the figure that both $K(\phi)$ and σ_{pc} closely match each other. A scaling behaviour in the I–V characteristics (Eq. (13)) was observed from the typical behaviour of current from non-linear to linear nature with increase in filler loadings as compared to conductivity variation from the power law model (Eq. (2)).

4.7. D-shore hardness

D-shore hardness of PS–foliated graphite composite with respect to ϕ is shown in Fig. 10. The hardness of the composites is found to decrease sharply from 89.5 to 85 up to the percolation concentration of the filler. However, beyond the percolation threshold, the hardness decreases slowly to 79.5 for $\phi = 0.115$. It is evident that using foliated graphite as filler, the degradation in the hardness of the PS/FG composite is only 12% at the highest achieved conductivity of ca. 10^{-1} S/cm. The decrease in hardness with increase in the FG content may be due to the decreased particle-to-particle interaction among PS particles in the presence of graphite conducting channels in the composite.

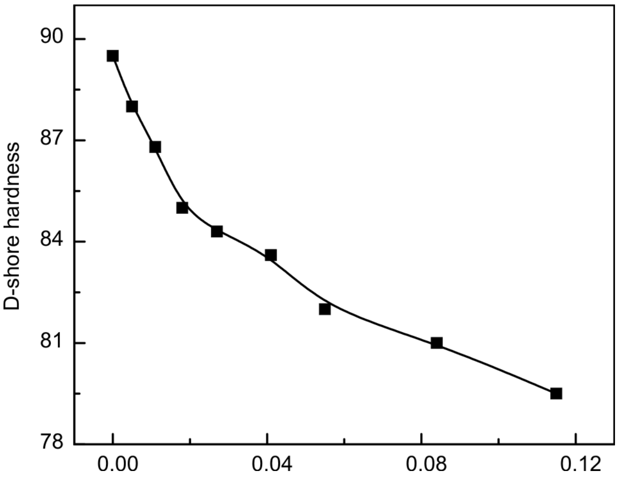

Fig. 10. D-shore hardness of PS/foliated graphite composites in a function of ϕ

5. Conclusion

The use of foliated graphite as filler was found to lower the percolation threshold. Electrical conductivity as high as 10^{-1} S/cm was obtained, without significantly affecting the hardness. The electrical conductivity data could be well understood in terms of the GEM equation. The dominant transport mechanism of charge carriers near and above the percolation threshold are found to be tunnelling and ohmic conduction, respectively.

Acknowledgement

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