Effect of Percolation on Electrical and Dielectric Properties of Acrylonitrile Butadiene Styrene/Graphite Composite

H. S. Dahiya,¹ Nawal Kishore,² R. M. Mehra³

¹Department of Physics, CRA College, Sonepat, Haryana, India

²Department of Applied Physics, GJ University, Hissar, Haryana, India

³Department of Electronic Šcience, University of Delhi South Campus, New Delhi 110 021, India

Received 17 February 2006; accepted 6 May 2007 DOI 10.1002/app.26896 Published online 23 July 2007 in Wiley InterScience (www.interscience.wiley.com).

ABSTRACT: Prelocalized Acrylonitrile Butadiene Styrene/graphite composites were prepared by hot compression molding technique. The increased conductivity with increase of graphite content exhibits percolation phenomenon. The current–voltage characteristics are found to change from nonlinear to linear above the percolation threshold. A positive temperature coefficient of resistance is observed in these composites, and this effect is more pronounced in samples having graphite concentration near percolation threshold. The dielectric constant was found to increase slowly up to the percolation concentration and beyond it a sudden increase in its value is observed. The dissipation factor exhibits maxima in the vicinity of percolation threshold. The dielectric properties are discussed in terms of the interfacial Maxwell-Wagner effects. © 2007 Wiley Periodicals, Inc. J Appl Polym Sci 106: 2101–2110, 2007

Key words: acrylonitrile butadiene styrene; graphite; composites; percolation; positive temperature coefficient of resistance; dielectric

INTRODUCTION

Filled electro-conducting polymer composites are suitable for a number of applications such as electromagnetic interference shielding in electronic devices, e.g., housings of computers and cellular phones, temperature and current sensor devices,^{1,2} current limiting devices in circuit protection,^{3–6} antistatic materials (chemicals which provide a conductive coating on a polymer surface so that static charges can be leaked off), self-regulating heaters, etc.^{7,8} One of the main areas of technical interest is to achieve conductivity at a relatively low filler concentration, as higher filler loadings tend to deteriorate the mechanical properties of the composites. The most popular method of predicting the properties of compo-sites is the percolation theory.⁹ It is based on the concept that an abrupt change in the properties will occur when the conducting phase makes a chain from one side to the other of the matrix polymer. The volume fraction at which this occurs is called

Journal of Applied Polymer Science, Vol. 106, 2101–2110 (2007) © 2007 Wiley Periodicals, Inc.



the percolation threshold. The percolation theory predicts a conductivity dependence of the form:

$$\sigma = \sigma_0 (v_f - v_{\rm fc})^t \tag{1}$$

where v_f is volume fraction of the conducting species, v_{fc} is the percolation threshold, i.e., the volume fraction below which the conductivity falls to a very small value, σ_0 is the conductivity of the conducting species, and *t* is an exponent determining the power of electrical conductivity increase above v_{fc} .

This article reports the change in conductivity of prelocalized graphite (Gr) filled Acrylonitrile Butadiene Styrene (ABS) composite as a function of graphite content. The data obtained are analyzed in terms of the percolation theory. The variation of resistivity with temperature for samples having different graphite contents has been studied to explore the possibility of utilizing these composites for making temperature-sensing devices. The current-voltage (I-V) characteristics are studied for understanding the conduction mechanism in these composites. The variation of dielectric constant, the dissipation factor, and the loss factor with the frequency of the measuring field as well as graphite content is also studied at ambient temperature to examine the effect of percolation on these parameters. The dielectric behavior of filled conducting composites is subject to interfacial polarization for heterogeneous systems when

Correspondence to: R. M. Mehra (rammehra2003@yahoo. com).

Contract grant sponsor: University Grants Commission (UGC), New Delhi, India.

 TABLE I

 Properties of Acrylonitrile Butadiene Styrene (ABS)

Property	Value
Color	Milky white
Density (g/cm^3)	1.12
Melt flow (g/10 min)	4.5
Hardness, Shore D	85
Electrical resistivity	
(at room temperature, RT) (Ohm-cm)	$1.849 imes 10^{11}$
Dielectric constant (at 100 kHz, RT)	2.96
Dissipation factor (at 100 kHz, RT)	0.00652
Glass transition temperature (°C)	107-115

one of the components of the composite has a higher electrical resistivity as compared with the other.¹⁰ This is also popularly known as Maxwell Wagner Effect. Over the years, a number of theoretical and empirical relations¹¹ for predicting the dielectric constant has been proposed but most of these fit better for dilute composites. D-Shore hardness as a function of graphite content is also measured to evaluate the mechanical strength of ABS/Gr composites

EXPERIMENTAL

The matrix polymer used in this work is a commercial grade (920) natural ABS resin manufactured by Lanxess ABS, Vadodara, Gujrat, India, supplied in the form of granules. A thermoplastic ABS matrix was selected because of its strength, durability, and relatively easier processibility. ABS is an amorphous, phase-separated polymer. Some of the properties of this resin are listed in Table I.

The electrical conducting filler is natural flake graphite supplied by Graphite India, Banglore, India. The scanning electron micrograph (SEM) of the graphite powder is shown in Figure 1. The graphite particles are flaky with particle size ranging from 10 to 20 μ m. Some properties of graphite are given in Table II.

The ABS granules were grinded to obtain finer particles (212-250 µm) using Aurther H. Thomas type Willey Grinder. The polymer and filler powders were tumble mixed thoroughly for 8 h in a glass beaker with the help of a magnetic stirrer. During this process, because of their sticky nature, the graphite particles get coated on to the surfaces of ABS particles. This process is referred to as prelocalization of the conductive phase. Prolong mixing improves the homogeneity of the spatial distribution of the conductive particles and their more uniform coating on polymer particles resulting in a low percolation threshold. The graphite coated ABS particles were compacted by compression molding at 90°C under a pressure of 105 MPa using a hydraulic press (Shimadzu Corp. Kyoto, Japan) having ram diameter 42.7 mm and mounted with a pressure gauge in a piston-cylinder assembly. The temperature and pressure were maintained for 15 min. All the prepared samples were then baked at 100°C, under 35 MPa pressure for 15 min.

To ensure uniform heating during sample preparation a hollow cylindrical heating filament was designed. A cleavage on the cylindrical surface was given and two screws were provided onto the two sets of clamps protruding from the surface of the metallic housing of the filament in the vicinity of the cleavage. These two screws helped in fixing or removing the heating filament, on or from the pressure cell. A small hole was provided in the molders as well as filament holder for inserting the thermocouple.

Three series of specimens of ABS/Gr conductive composites with different contents of graphite were prepared. Disk shaped specimens of diameter ~ 1 cm and thickness ranging from 0.3 to 0.6 cm were prepared. The surfaces of the specimens were polished with emery paper to remove the polymer-rich surface layer and to eliminate surface irregularities. All of the specimens were sealed in air free polyethylene bags prior to testing to avoid atmospheric and humidity effects that may induce some changes in the surface conductivity of the samples. Both surfaces of the sample were painted with conductive silver paint. The copper electrodes were bonded to samples with conductive silver paint.

The resistance of the samples having different graphite contents was measured at room temperature using digital multimeter when the resistance was less than 200 M Ω . For higher resistances a picoammeter was used. The dc conductivity of the samples was then calculated using the relation:

$$\sigma = \frac{d}{RA} \tag{2}$$



Figure 1 SEM image of the graphite powder.

TABLE II Properties of Graphite

Property	Value
Density (gm/cm ³) Color Modulus of elasticity (Young's modulus) (GPa) Max. use temperature (inert atmosphere) (°C) Electrical resistivity (at RT) (Ohm-cm)	$\begin{array}{c} 1.75 \\ \text{Black} \\ 4.8 \\ 3600 \\ 7.5 \times 10^{-5} \end{array}$

where "*A*" is cross-sectional area and "*d*" is the thickness of the sample. The resistivity (ρ) of the samples was then determined according to the relation: $\rho = 1/\sigma$. The resistivity as a function of composition has been studied.

The current–voltage studies were conducted with a Keithley Sourcemeter 2400 attached to a computer to access and store data directly. A dc excitation with in the voltage range of 0–50 V was applied to the samples at room temperature.

To explore the possibility of utilizing the composite material for temperature sensor, the effect of temperature on resistivity was studied. For study of positive temperature coefficient of resistivity (PTCR) phenomenon, sample holder made of brass having large thermal capacity and a proper arrangement for holding sample as well as thermocouple has been used. The wired samples were placed in the sample holder having proper arrangement for thermal insulation. A Cr/Al thermocouple connected to a digital temperature indicator (Autonix, PP-35) was placed in direct contact with the samples to measure the temperature. A heating rate of 2°C/min was maintained to give enough time to the sample to absorb heat from the surroundings.

The permittivity (ε') and dielectric loss (ε'') of the samples were determined at frequencies ranging from 10 kHz to 10 MHz using a Hewlet Packard Impedance Analyzer (model No. 4294). The capacitance (*C*) and the loss tangent (tan δ) were measured directly. ε' and ε'' were calculated using the expressions:

$$\varepsilon' = \frac{Cd}{\varepsilon_0 A} \tag{3}$$

$$\varepsilon'' = \varepsilon' \tan \delta$$
 (4)

where ε_0 is the permittivity of the free space.

All the measurements were done perpendicular to the flat faces of the samples under ambient conditions. Scientico Hardness tester (Durometer, model No. SRHT-501D) conforming to ASTM 1706-61 and D 676-59T specifications is used to determine the hardness of the samples.

RESULTS AND DISCUSSION

The conductivity versus composition—The power law

Figure 2 shows the variation of conductivity as a function of graphite volume fraction. At the onset of percolation the conductivity increases of nearly eight orders of magnitude is observed within a narrow range of graphite concentration.

Equation (1) can be rewritten as

$$\log \sigma = \log \sigma_0 + t \log \left(v_f - v_{\rm fc} \right) \tag{5}$$

Figure 3 shows the relationship between log σ and $\log (v_f - v_{fc})$ for the ABS/Gr composite system. Linear relationship between log σ and log ($v_f - v_{fc}$) is observed. The best linear fit has been obtained for $v_{fc} = 0.0252$ which is in good agreement with the experimental value of 0.029. The small difference in the two values may be attributed to the limitation to take a finite number of experimental points. The values of critical exponent t and σ_0 as calculated from the slope and intercept are found to be 3.37 and 1.35 \times 10⁴ S/cm, respectively. The value of σ_0 is close to the value of the conductivity of the filler graphite $(1.33 \times 10^4 \text{ S/cm})$. According to Kirkpatrick⁹ the value of t is 1.5 ± 0.2 while Straley¹² found t to be equal to 1.75 ± 0.1 for a 3D system. However, Wang and Rubner¹³ have reported a value of t = 3.2 for their system.

I-V characteristics

The nonlinear current–voltage behavior is one of the electrical characteristics for most of the conducting polymer composite materials. The I-V characteristics of most filled polymers satisfy the following equation:



Figure 2 Conductivity as a function of graphite volume fraction.

Journal of Applied Polymer Science DOI 10.1002/app

0 -1 -og σ_m(S/cm) -2 -3 -4 -5 -2.0 -1.6 -1.2 -2.4 $Log (v_f - v_{fc})$

Figure 3 Log of conductivity as a function of log (v_f – v_{fc}).

$$I = CV^m \tag{6}$$

where V stands for the applied voltage, I, the corresponding current, C is the conductance and "m" is the slope of the log $I - \log V$ plot. Figure 4(a-e) shows the I-V characteristics of the prelocalized ABS/Gr composite with different graphite contents. The variation of "m" with the graphite volume fraction is shown in Figure 4(f). It is clear from the figures that the I-V behavior of the prelocalized composites is dependent on graphite content. When m = 1, we have the ohmic behavior. The above equation can be rewritten as

$$\log I = m \log V + \log C \tag{7}$$

The log $I - \log V$ behavior of the corresponding samples is shown as inserts in these figures. The composites with high content of graphite exhibit value of m close to 1. In the vicinity of the percolation threshold a sharp increase in the value of "m" is observed and its value becomes 1.481 for the sample having minimum value of graphite volume fraction, i.e., 0.024.

Mamunya et al. and Feng and Chan^{14,15} in PE/ POM-Fe composites have also observed a decrease in the value of "m" with the increase of the volume fraction of the conductive filler. Thus the percolation threshold may also be assumed as that value of volume fraction of the filler where the transition from nonohmic to ohmic current takes place.

Various mechanisms have been proposed to explain this nonlinear electrical behavior in conducting polymer composites. The reason of nonlinearity of the I-V characteristics in the PP-Ni polymer metal composite according to Chen and Johnson¹⁶ is the creation of strong local electric fields between the conductive clusters.

Mainly, the following two mechanisms of charge transport can exist in composites loaded with conducting fillers.

- 1. Ohmic conductivity through direct contact of filler particles.
- 2. Nonohmic conductivity by means barrier tunneling between filler particles, which have no direct contact.

Decrease in interparticle distances as well as the number of tunneling gaps causes the transition. The transition of the conduction mechanism from nonohmic to ohmic is thus expected in the percolation transition. The filler concentration is an important parameter in determining the network microstructure in a given composite system. As v_f approaches v_{fc} from below enough particles begin to come closer decreasing tunneling gaps significantly and a network of conducting channels is formed. Within the percolation transition region the particle clusters are joined together in a bond percolation model that governs the conduction. Polymer layers inevitably exist between the clusters and when the gaps among the clusters are small enough, electron tunneling occurs readily. At sufficiently high filler content, for $v_f > v_{fc}$ agglomeration of the clusters takes place and an infinite percolation network generates resulting into ohmic conduction.

The resistivity-temperature study

Figure 5(a-d) shows the resistivity of the ABS/Gr composite system as a function of temperature. The resistivity increases with the increase in temperature for all the four graphite concentrations examined. These results indicate that the positive temperature coefficient of resistance (PTCR) is dependent on the graphite concentration. The ratio (R_P/R_{RT}) is found to be greater than 100 for 2.9 vol % of graphite composite, ranges from 4 to 5 for 3.6 and 4.9 vol % of graphite composites and is about 2.4 for 7.6 vol % of graphite composite, where $R_{\rm P}$ and $R_{\rm RT}$ are peak and room temperature resistivities, respectively. Thus as the graphite concentration decreases and approaches v_{fc} from above the PTCR effect increases. Good reproducibility was found on repeated heating and cooling cycles although a slight increase in the RT resistivity value is noticed after every cycle. The maximum change of resistivity (~ 100 times) occurs for 2.9 vol % of graphite.

When the composite is heated a few degrees beyond 130°C, the resistivity further nominally increases but on cooling the resistivity value do not





Figure 4 Variation of current with voltage for (a) 2.4, (b) 2.9, (c) 3.6, (d) 4.9, (e) 7.6 vol % graphite, and (f) variation of "*m*" with graphite vol %.

come down to its initial value. As the temperature increases a gradual increase in the resistivity is attributed to the differences in the coefficients of expansion of the filler and the polymer matrix, which causes an increase in the interparticle distance of filler particles. The expansion of the polymer with temperature causes a decrease in the number of conductive graphite channels. This is because some of the thin graphite channels may break as the polymer expands resulting in the reduction of conductive paths. It is for this reason that a larger PTCR effect is observed for composite systems having low

Journal of Applied Polymer Science DOI 10.1002/app



Figure 5 The resistivity of ABS/Gr composite as a function of temperature with (a) 2.9, (b) 3.6, (c) 4.9, and (d) 7.6 vol % graphite.

graphite content, as the dimensions of the conducting filler paths in these materials are small. Such a dramatic reduction in the number of conductive paths would not occur in the high graphite content composites systems in which the dimensions of the graphite channels are large. In this case, the polymer expansion would only decrease the dimensions of conductive paths but would not break them up. This observation is reflected in Figure 5(d), which shows the resistivity as a function of temperature for the composite material containing 7.6 vol % of graphite. This plot shows that only a small change in the resistivity is observed beyond T_g of the matrix polymer and that the change is much smaller in comparison to the change in the resistivity of the composite system containing 2.9 vol % of graphite shown in Figure 5(a). An important observance that can be made from the present study is that the abrupt change in the resistivity occurs in the same temperature range irrespective of the graphite content in the composite and this temperature range is just above

the glass transition temperature range of the polymer matrix. This effect is more pronounced in samples having graphite concentration near percolation threshold.

These results support the theory that amorphous polymers have two thermal expansion coefficients, one of them, β_g , to be independent of temperature at $T < T_g$ and second, β_{l} to be weakly dependent on temperature at $T > T_g$ and greater than the first.¹⁷

The dielectric studies

The total complex permittivity or the complex dielectric constant (ε^*) can be represented as

$$\varepsilon^*(\omega) = \varepsilon'(\omega) - i\varepsilon''(\omega) \tag{8}$$

where $i = \sqrt{-1}$ and $\omega = 2\pi f$ is angular frequency of the measuring electric field. The real part of the relative permittivity, ε' , commonly known as the



Figure 6 Variation of dielectric constant with (a) graphite content and (b) frequency.

"dielectric constant" is associated with the polarization or capacitance of the material and the imaginary part, ε ", generally called the "loss factor" or "dielectric loss" is associated with its conductance. The ratio of the imaginary to the real parts (ε "/ ε ') is called the dissipation factor, represented by tan δ , where δ is the angle between the voltage and the charging current. This angle δ is called the "loss angle".¹⁸

Figures 6–8 show the dielectric properties obtained for ABS/Gr composite systems. The variation of dielectric constant with filler concentration is presented in Figure 6(a). The measurements taken at four different frequencies, i.e., 10 kHz, 100 kHz, 1 MHz, and 10 MHz are shown. At 10 kHz the value of dielectric constant is 4.89, 5.71, and 31.37 for 2.4, 2.9, and 3.6 vol % of graphite samples, respectively. Similar behavior is observed for higher frequencies also. The dielectric constant thus increases moderately up to a point in the vicinity of the percolation threshold obtained from the resistivity concentration curves (Fig. 2). Above this point the dielectric constant increases at a much faster rate and exhibit an abrupt change.^{19,20} The permittivity increases due to space charge built up at the interfaces between the conducting phase and the insulating phase due to difference in the conductivity of two phases.²¹ Space charge polarization occurs when more than one material component is present or when segregation occurs in a material containing incompatible chemical sequences and when translating charge carriers become trapped at the interfaces of these heterogeneous systems. The electric field distortion caused by the accumulation of these charges increases the overall capacitance of a material, which appears as an increase in the dielectric constant. In the present study, also the relative mobility of the charge carriers in ABS and graphite results in the carriers being trapped in the interfaces of separation.

Bergman and $Imry^{22}$ and Wilkinson et al.,²³ by computer simulation using models based upon ran-



Figure 7 Variation of loss factor with (a) graphite content and (b) frequency.



Figure 8 Variation of dissipation factor with (a) graphite content and (b) frequency.

dom distributions of resistors and capacitors have shown that dielectric constant increases which increase as the percolation threshold is approached from either side. However, in the present work the dielectric constant is found to increase with the increase in filler concentration even after the percolation threshold. In fact above the percolation threshold (i.e., when the samples become conductive) a rapid increase in the value of the dielectric constant is observed and beyond this point the data could not be measured because of the large increase in the conductivity of the composites. A plausible explanation for this type of behavior may be that as the graphite content increases the interparticle distances decrease and local fields created by neighboring particles enhance the rise in the value of the dielectric constant. At lower field frequencies this effect is more pronounced.

Steeman¹¹ derived the following equation for predicting the dielectric constant of the composite.

$$\varepsilon_c = \varepsilon_m \left(\frac{1 + 2v_f}{1 - v_f} \right) \tag{9}$$

where ε_{c_i} is the dielectric constant of the composite, ε_m is the dielectric constant of the unfilled polymer matrix at a given frequency, and v_f is the filler volume fraction. In Figure 6(a) the data obtained by using eq. (9) at 10 kHz is also plotted for a comparison. The data however show consistency with the experimental data only for low concentrations of graphite. As the graphite concentration approaches threshold concentration, strong deviations are observed.

Figure 6(b) shows the variation of dielectric constant with the frequency of the applied field, ranging from 10 kHz to 10 MHz. The dielectric constant, ϵ' of the unfilled polymer matrix and of other samples below the percolation threshold concentra-

Journal of Applied Polymer Science DOI 10.1002/app

tion (2.9 vol %) is found to vary from 2 to 6 and exhibit a moderate decrease with the increase in frequency. For 3.6 vol % of graphite composite, however, the value of ε' falls from 31.7 to 9.5 when the frequency increases from 10.7 kHz to approximately 10 MHz. Thus above the percolation threshold a pronounced fall in the value of ε' with increasing frequency is observed. The value of ε' is higher at lower frequencies, as a consequence of the interfacial Maxwell Wagner Polarization effect between the polymer and graphite.²⁴ The value of ε' decreases with increasing frequency because the dipoles orient with some delay thanks to their own relaxation time.²⁵

The variation of loss factor, ε'' , with composition is shown in Figure 7(a). At 10 kHz its value increases from 0.018 to 22.86 as the graphite content increases from 1.4 to 3.6 vol %. The value of ε'' appears to increase with increasing graphite content in a manner observed with the dielectric constant values of Figure 6(a), though the change in the losses in the composites is more dramatic than in their corresponding dielectric constant values. The losses too thus increase with the increase in graphite concentration within the frequency ranges studied in accordance with the Maxwell-Wagner polarization theory. The percolation threshold is clearly discernible in both ε' and ε'' versus graphite concentration plots. The addition of conductive filler into the polymer beyond critical concentration thus increases the dielectric constant enormously. However as the composite becomes conductive, the loss factor increases and the material cannot operate efficiently as a high dielectric constant material.

Figure 7(b) shows the loss factor, ε'' , as a function of frequency for different graphite contents in the composite. The values of ε'' at 10.7 kHz for 2.4, 2.9, and 3.6 vol % of graphite samples are 0.08, 0.206, and 22.86, respectively. The corresponding values of ε'' at 10 MHz are 0.15, 0.207, and 1.55. Thus a

decrease in the value of ε'' for the conducting and an increase for the insulating samples with increasing frequency is observed. The samples having graphite content near percolation threshold exhibited a more or less frequency independent behavior.

The variation of dissipation factor with graphite content at different field frequencies is shown in Figure 8(a). At 10.7 kHz the value of dissipation factor is 0.0055 for 1.4 vol % of graphite sample. The dissipation factor, tan δ , moderately increases with the graphite content roughly up to the percolation threshold concentration by the interfacial polarization mechanism similar to that of the dielectric constant. As the graphite loading increases and surpasses v_{fc} an abrupt rise in the value of the dissipation factor is observed and it attains a value of 1394.29 at 10.7 kHz. A further increase in the graphite concentration causes a decrease in the concentration of the induced dipoles due to the formation of a network of conductive channels, leading to a decrease in the dissipation factor value. Despite a fall in the dissipation factor, its value is still high (88.126) in comparison to that of the unfilled polymer (0.00487 at 10.7 kHz), the maximum tan δ values are roughly associated with the percolation threshold. The high values of the dissipation factor obtained for these composite systems may be attributed to the skin effect associated with a relatively high penetration level of electrical field into graphite particles.

In Figure 8(b), the dependence of dissipation factor on the frequency of the measuring field is shown. A behavior similar to that of the loss factor is observed. At 3.6 vol % and higher graphite content, i.e., above v_{fc} , a decrease in the value of tan δ is observed. The most pronounced fall in the value of tan δ is observed for the 4.9 vol % of graphite composite where an increase in frequency from 10.7 kHz to 10 MHz causes a decrease in its value from 394.28 to 2.4. The samples having graphite content below percolation threshold exhibits an increasing trend with the increase in frequency. Generally, the magnitude of dissipation factor, as well as other dielectric properties, is sensitive to the polar sites in the polymer matrix. These sites increase the dissipation factor and may confuse the dependence of tan δ on frequency. For a given concentration above the critical concentration, the dissipation factor decreases with an increasing frequency because the induced charges gradually fail to follow the reversing field causing a reduction in the electronic oscillation as the frequency is increased.

Hardness

Figure 9 shows the D-shore Hardness as a function of graphite content. A pronounced decrease in the

Figure 9 Variation of D-Shore Hardness with graphite vol %.

hardness with initial increase in graphite content roughly up to the percolation concentration and a relatively small decrease thereafter are observed. This is suggestive of a decreased adhesive force between the ABS particles due to the presence of graphite particles in the interfacial regions.

CONCLUSIONS

The observed percolation threshold in ABS/Gr composites is found to have a phenomenal impact on I-V characteristics, the dielectric behavior, and the hardness. I-V characteristics are found to change from linear to nonlinear in the vicinity of the percolation threshold. The composites possess the PTCR effect, which decreases with increase of filler content and the maximum change in resistivity is observed for the composite having filler content in the vicinity of percolation threshold. In fact for higher loadings of graphite there is a nominal change in resistivity with in the temperature range studied. Good repeatability was found on repeated heating and cooling of the composite when heated up to 130°C, a temperature slightly higher than glass transition temperature of ABS, although a slight increase in the RT resistivity value is noticed after every heating cycle. Since the maximum change in resistivity has occurred for 2.9 vol % of graphite composite in the same temperature range, this composite having graphite concentration near the percolation concentration is suitable for device applications.

The dielectric constant and the loss factor of ABS/ Gr composite are found to be strongly dependent on the graphite content at and above percolation threshold exhibiting a sharp increase in their value. Below the critical concentration, i.e., the percolation threshold, the interparticle (filler) distance is large enough



so that local fields created by apparently neighboring particles do not interact. Thus, as observed, the dielectric constant in this concentration region increases slowly. As the filler concentration increases the Maxwell Wagner effect increases leading to a rise in the values of dielectric constant. Above the critical concentration the composite contains a network of continuous chains and the concentration of inductive charges is reduced. This causes a decrease in the dissipation factor. The dielectric constant of the composite above the critical concentration is found to be higher at lower frequencies, which could also be a consequence of Maxwell Wagner polarization. The loss factor and the dissipation factor are found to increase with frequency below percolation threshold while above percolation threshold, a decrease in their value is observed.

References

- 1. Strumpler, R. J Appl Phys 1996, 80, 6091.
- 2. Ploss, B.; Shin, F. G.; Chan, H. L. W.; Choy, C. L. Appl Phys Lett 2000, 76, 2776.
- 3. Strumpler, R.; Maidorn, G.; Rhyner, J. J Appl Phys 1997, 81, 6786.

- 4. Doljack, F. A. IEEE Trans Comput Hybrids Manuf Tech 1981, CHMT-4, 372.
- 5. Dougal, R. A. IEEE Trans Power Electron 1996, 11, 304.
- 6. Duggal, A. R.; Levinson, L. M. Appl Phys Lett 1939 1997, 71.
- 7. Kimura, T.; Yasuda, S. Polymer 1988, 29, 524.
- 8. Kimura, T.; Asano, Y.; Yasuda, S. Polymer 1996, 37, 2981.
- 9. Kirkpatrick, S. Rev Mod Phys 1973, 45, 574.
- 10. Meakins, R. J Prog Dielectrics 1961, 3, 151.
- 11. Steeman, P. A. M. Ph. D. Thesis, Delft University 1992.
- 12. Straley, J. Phys Rev B 1997, 15, 5733.
- 13. Wang, Y.; Rubner, M. F. Macromolecules 1992, 25, 3284.
- Mamunya, Y. P.; Muzychenko, Y. V.; Pissis, P.; Lebedev, E. V.; Shut, M. I. Polym Eng Sci 2002, 90, 42.
- 15. Feng, J.; Chan, C. M. Polym Eng Sci 1998, 38, 1649.
- 16. Chen L. G.; Johnson, W. B. J Mater Sci 1995, 26, 1565.
- 17. Perepechko, I. I. An Introduction to Polymer Physics. (Beknezazarovs, A., Translator); Mir Publishers: Moscow 1981, Chapter 4.
- 18. Coelho, R. Physics of Dielectrics for the Engineers; Elsevier: Amsterdam, 1976.
- 19. Narkis, M.; Vaxman, A. J Appl Polym Sci 1984, 29, 1639.
- 20. Yacubowicz, J.; Narkis, M. Polym Eng Sci 1986, 26, 1568.
- 21. Hippel, A. R. V. Dielectrics and Waves; Wiley: New York, 1953.
- 22. Bergman, D. J.; Imry, Y. Phys Rev B 1977, 39, 1222.
- 23. Wilkinson, D.; Langer, J. S.; Sen, P. N. Phys Rev B 1983, 28, 1081.
- 24. Benguigui, L.; Yacubowicz, J.; Narkis, M. J Polym Sci Polym Phys Ed 1987, 25, 127.
- 25. Svorcik, V.; Gardasova, R.; Rybka, V.; Hnatowicz, V.; Cervena, J.; Plesek, J. J Appl Polym Sci 2004, 91, 40.