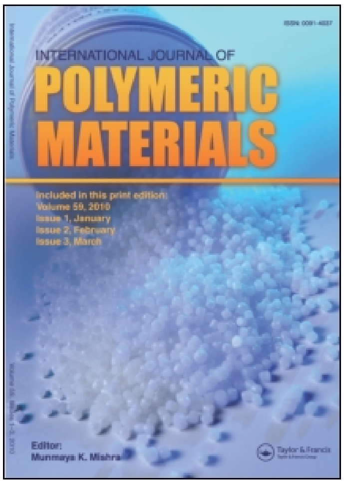

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The Electrical Behavior of Mica-Polystyrene Composite

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The electrical properties of mica-polystyrene composites were studied using the impedance measurements technique. The study was carried out as a function of frequency and mica concentration. It was observed that the AC conductivity and the dielectric constant are increased with increasing of the mica content in the composite. The observed electrical results fit approximately the reported empirical equations concerning the AC conductivity and dielectric behavior of polymer composites. Relaxation processes were observed to take place for composites having high mica concentration. The observed relaxation and polarization effects of the composite are mainly attributed to the dielectric behavior of the mica filler. However, the results were explained on the basis of the interfacial (space-charge) polarization, dipolar polarization and decrease of the hindrance produced by the polymer matrix.

Keywords: Electrical properties; mica; polystyrene; composite; impedance; frequency; dielectric constant; conductivity; polarization

1. INTRODUCTION

Polymer composites are recent advanced materials appeared in the last twenty years and have numerous important technological applications. The composite consists of a filler as a reinforcement element and a surrounding binder or matrix fabricated into a useful product. Filled-reinforced polymer composites have gained a large area of application due to their good durability, technical performance and

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enhanced electrical properties. One application of the conductive composities is their uses in electrostatic charge protection and electromagnetic interference shielding [1 – 3].

The addition of mineral fillers in polymers become in the last three decades very popular research interest to improve the mechanical, electrical, thermal, optical, and processing properties of polymers. Consequently, the mineral filled polymer composites have become widely accepted in electrical and other applications. Mica is an attractive candidate as a filler in electrical applications because it has excellent insulating properties. Mica, when used as a reinforcing agent in plastics, offers many advantages including planer reinforcement, reduced creep, *etc.* The study of the effect of mica on the electrical properties of polystyrene is important from the practical point of view, since polystyrene is extensively used as an electric insulator in many industrial applications [4 – 6].

The polymer composite used in the present study is mica/polystyrene with different mica concentration. The mica filler is muscovite mineral of chemical composition $K_2Al_4(Al_2Si_6O_{20})(OH)_4$ obtained from natural rocks in Jordan. Mica is a dipolar material having low dielectric loss and dielectric constant in the range 6 – 8. The polystyrene matrix is characterized by low cost, case-processing, hardness, and good dielectric properties. The properties of this nonpolar can be tailored to provide good mechanical, thermal, and chemical properties [1, 7].

The main objective of this study is giving information concerning the electrical behavior of mica/polystyrene composite using the impedance spectroscopy which is one of the powerful techniques to characterize the dielectric properties as we reported in several previous publications [9 – 12]. Therefore, thin films based on polystyrene with natural mica as a reinforcement filler were used in the present study. The effects of the mica concentration and the frequency of the applied field in the range (300 Hz – 10 kHz) on the electrical properties of the prepared composites such as impedances, phase angle, dielectric constant, dielectric loss, and AC conductivity were studied. The morphology of the films was also investigated by SEM.

2. EXPERIMENTAL

2.1. Composite Preparation

One way to prepare polymer composites is casting from solution. It means simply dissolving the polymer by a solvent and pouring the solution into a mold and solidified *in situ* [5]. The natural mica used in this study was of sheet form. The mica filler was ground into powder by Agate mortar and sieved by U.S. standard sieve of size (63 μm).

Pre-weighed amount of polystyrene was dissolved in carbon tetrachloride CCl_4 at 70°C. Mica loaded thin films were prepared by dispersing mica powder in PS solution in different weight ratio. The solution was poured onto a stainless steel ring of a diameter 3.5 cm resting on a teflon substrate and allow evaporating the solvent at 65°C under atmospheric pressure. After solidification the specimens were easily detached from the teflon substrate. The thickness of the obtained specimens is shown in Table (I).

2.2. Scanning Electron Microscopy

To examine both the grain size and surface morphology of the composite specimens, Leitz scanning electron microscope (AMR, model 1000 A) was used. Figure (1) shows SEM micrographs for two mica concentration of composite samples of 10 and 50 wt.% of mica loaded in polystyrene matrix. These micrographs indicate the distribution of mica grains with no adjacent contacts between them. The

TABLE I The prepared composite samples

Specimen wt.%	*Thickness (μm)*
00	280
05	270
10	290
15	303
30	380
50	450
70	400

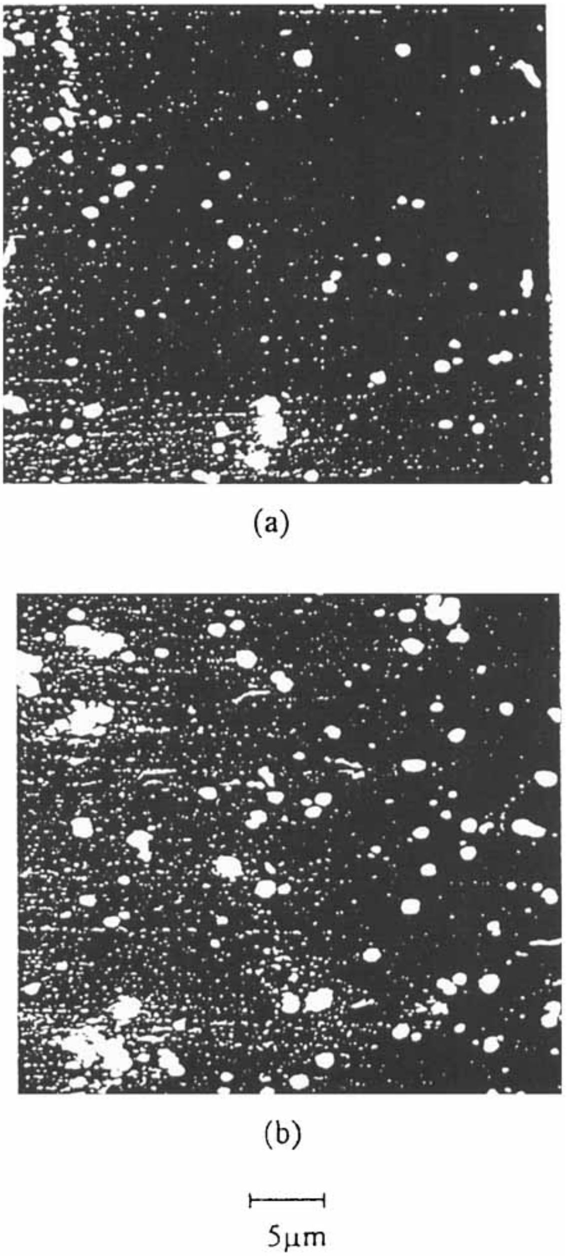

FIGURE 1 SEM micrographs for: (a) 10 wt.% mica content; (b) 50 wt.% mica content.

average grain size of mica powder was calculated and found to be in the range 5–40 μm.

2.3. Impedance Measurements

The phase shift and the AC-impedance of the given composite were measured by a gain-phase meter. This instrument is capable of measuring directly the ratio of the input to the output signals in dB and the phase angle ϕ in degrees as a function of frequency. The real component (ε') and the imaginary component (ε'') of the complex dielectric constant (ε^*) are related to impedance (Z) and phase angle (ϕ) as:

$$\varepsilon' = Z_c/2\pi f C_0 Z^2 \tag{1}$$

$$\varepsilon'' = Z_r/2\pi f C_0 Z^2 \tag{2}$$

where, f is the frequency, $C_0 = \varepsilon_0 A/T$ is the electrodes capacitance, T the specimen thickness, ε_0 the permitivity of free space, A the area of the disk, Z_c and Z_r are the imaginary and real components of the complex impedance ($Z = Z_r - jZ_c$), respectively [8,9]. The AC electrical conductivity (σ) was calculated from the relation:

$$\sigma = 2\pi f \varepsilon_0 \varepsilon'' \tag{3}$$

3. RESULTS AND DISCUSSION

In spite of the effect of mica filler on the dielectric relaxation of polar PVC films reported by Tripathi *et al.* [13], we feel our study is something else from the point view that the used polystyrene matrix is nonpolar and the mica filler is a polar natural material. Further more, we attempted in this study to analyze the observed electrical results by fitting some reported empirical equations concerning the AC conductivity and dielectric behavior of polymer composites [4–16].

All impedance measurements were taken at room temperature in the frequency range 300 Hz – 10 kHz. The composite specimens contain different mica concentration as shown in Table (I). The low frequency (less than 300 Hz) results were neglected to avoid the effect of electrode polarization. It was observed that the measured phase angles have always negative values for all samples of different mica concentration, which indicate that the composite consists of capacitive and resistive (series and parallel) networks. The movement of φ towards higher negative values with the increase of mica content indicates that the composites become more resistive than capacitive. This may be attributed to the existence of leakage current in the bulk composite, which increases with mica content, or may be attributed to hopping of ions by tunneling through the mica grains facilitated by the decreasing of the interdistance between the grains as the concentration is increased [12, 17].

Figure (2) shows the dependence of the impedance (per unit length) on the mica concentration at different frequencies (300 Hz, 1 kHz and 10 kHz) for the prepared composite films. Generally, it can be seen that

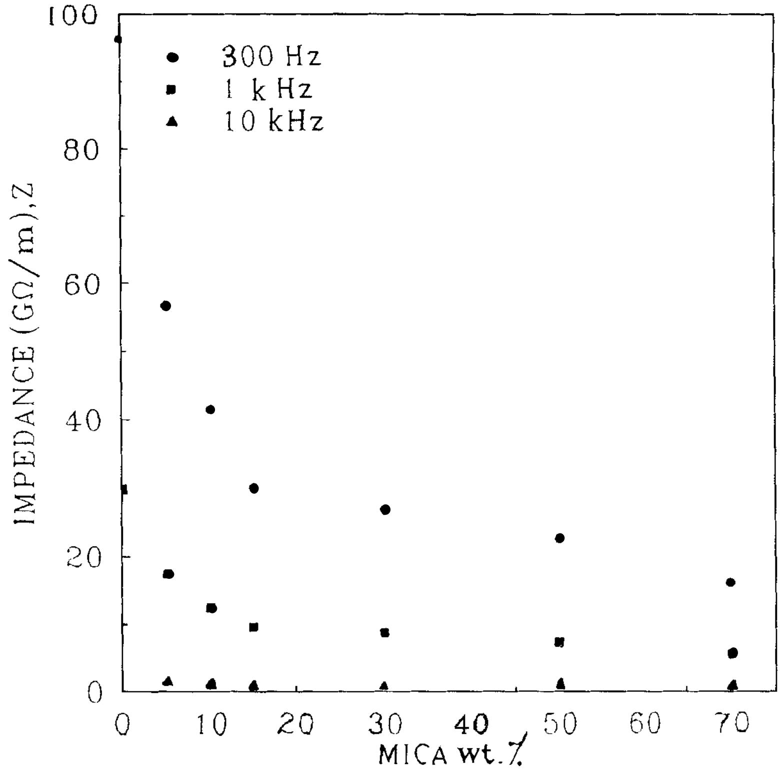

FIGURE 2 The impedance variation against the mica content.

the impedance decreases with increasing the mica content. This decrease in impedance is due to both the increase of mica concentration and the decrease of hindrance of polymer matrix [13, 18].

The variation of the impedance (per unit length) with frequency is shown in Figure (3). It was found that, for all the samples, the impedance at frequencies less than 1 kHz is relatively high and decreases exponentially with frequency. This may be due to the space charge in specimens or to some structural defects in addition to the electrode polarization effect [9 – 12]. However, above 1 kHz and up to 10 kHz, the impedance remains nearly constant. The dependence of the dielectric constant (ε′) on the frequency is shown in Figure (4). It was observed that the dielectric constant of all the samples is higher than the dielectric constant of the pure polystyrene, and for samples having

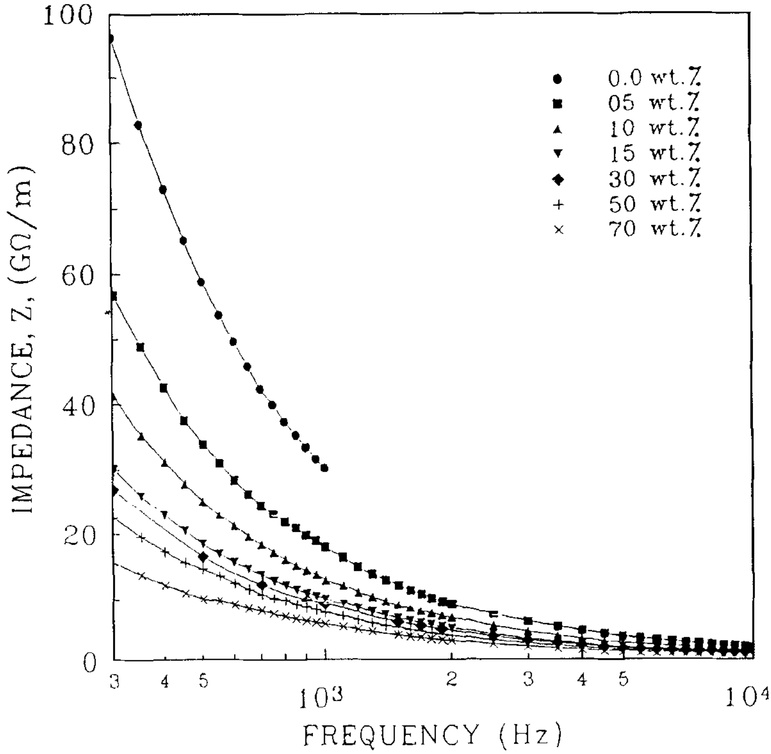

FIGURE 3 The variation of the impedance with the frequency.

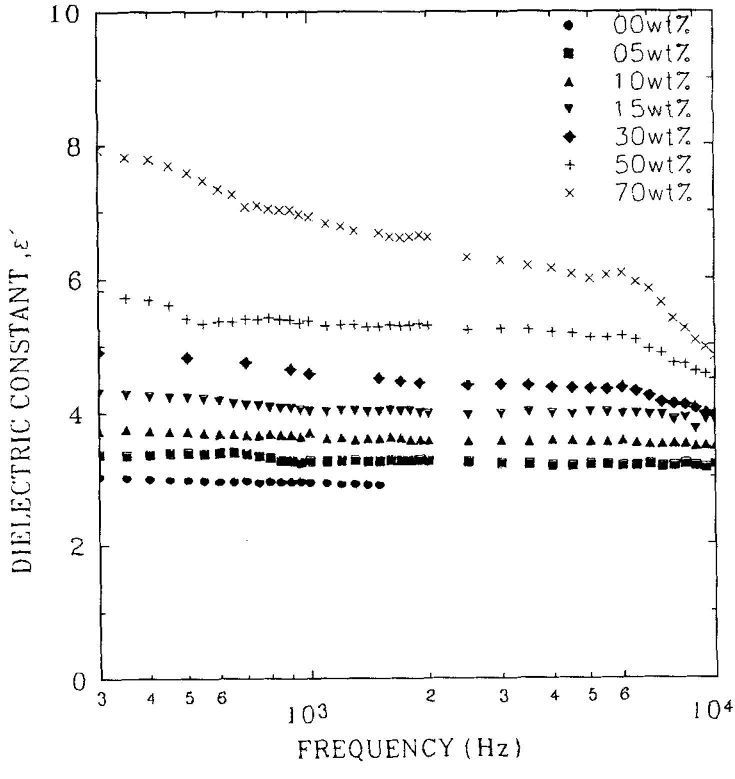

FIGURE 4 The frequency dependence of the dielectric constant.

concentration less than 15 wt.% is nearly independent of the frequency of the applied field. On the other hand, for samples having concentration higher than 15 wt.% a dispersion at low frequency is observed. This may be explained on the basis of space charge polarization produced from the mica grains which may act as trapping centers [10, 12, 19].

The behavior of the dielectric loss (ε'') against frequency is shown in Figure (5). The dielectric loss was found to be independent of the frequency for samples having concentration less than 5 wt.%. This independency is expected because polystyrene is a dominant component in the composite specimens with low mica content and it is a non-polar polymer. But for samples of higher mica concentration a dependence is observed especially with decreasing of the frequency.

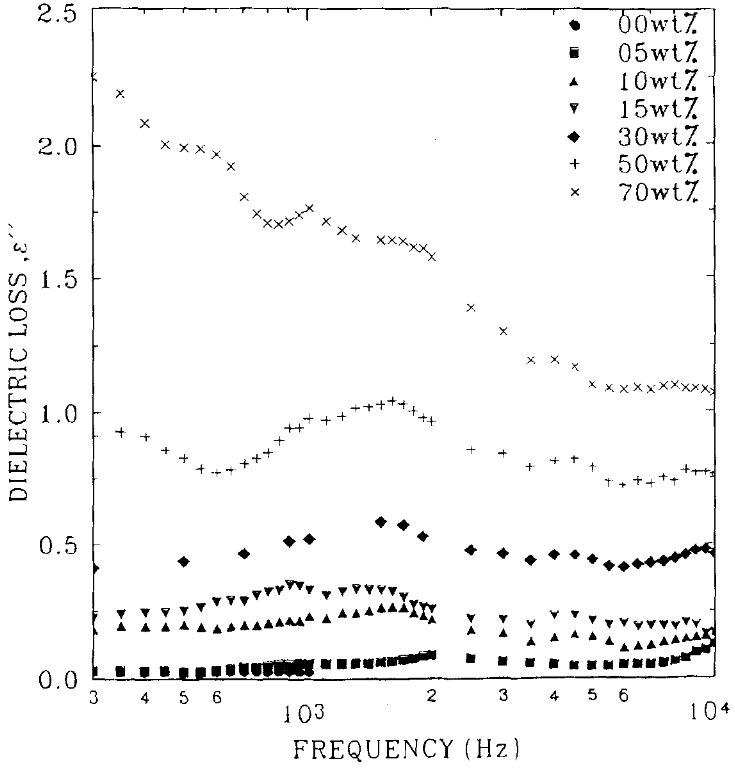

FIGURE 5 The variation of the dielectric loss as a function of frequency.

Also, a broad frequency peak was found in composite specimens of 10, 15, 30, 50 and 70 wt.%, which indicates a dipolar relaxation process coming from the constituents of the composite. On the other hand, Figure (5) shows another relaxation process occurs in 50 and 70 wt.% specimens at low frequencies, which is believed to be due to space charge polarization [21, 22].

The behavior of the AC conductivity is shown in Figure (6), where its value is very small and nearly constant below 700 Hz. But above 700 Hz and up to 1.5 kHz, the conductivity increases slowly for all the composite samples. At higher frequencies, *i.e.*, above 1.5 kHz and up to 10 kHz, the conductivity increases rapidly. This result supports the well known fact that the bulk AC conductivity is induced at high frequency range as it was observed by many researchers on different

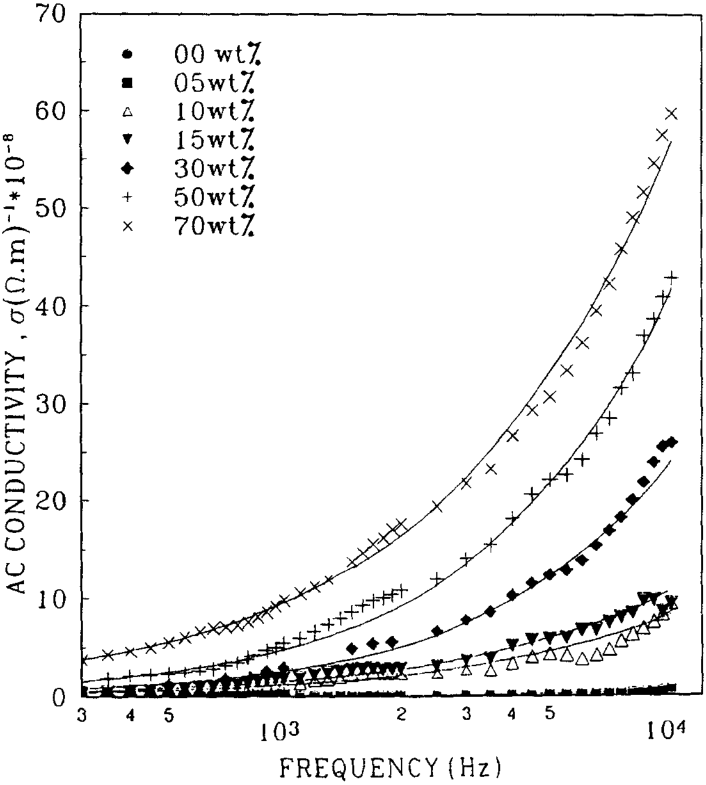

FIGURE 6 The AC conductivity of the composites as a function of frequency.

composite materials. This enhanced conductivity may be due to the increase of the electronic and ionic mobility of the existing impurities [9, 17]. Now, let us proceed on our discussion with further analysis and fitting the experimental data.

3.1. Cole–Cole Dielectric Analysis

Cole–Cole proposed a modification of the Debye equation [15, 19] which is applied for a single relaxation times as:

$$\varepsilon^* = \varepsilon_\infty + \frac{\varepsilon_s - \varepsilon_\infty}{1 + (i\omega\tau)^\beta}, \quad 0 < \beta < 1 \tag{4}$$

where, ε_s is the static dielectric constant, ε_∞ is the optical dielectric constant, τ is the relaxation time, ω is the frequency, and β is a measure of deviation from Debye equation. This Cole–Cole equation is frequently used as a successful tool to analyze the dielectric data of polymeric materials. The standard method to test the applicability of this model is *via* the so called Cole–Cole plot. A given point on this plot corresponds to a measurement of ε' and ε'' at a particular frequency. According to this model, plotting of ε' and ε'' for a normal dielectric material as polymers yields a semicircle. However, the geometry of the circle depends greatly on the polarity and structural uniformity of the dielectric materials. Generally, we can demonstrate the applicability of the Cole–Cole plots constructed for the given mica/PS composite using a computer program working at least squares fit. The parameters calculated from fitting the experimental points to assumed semicircles for composite samples of different mica concentration are shown in Table (II).

We can argue the Cole–Cole plots constructed for the given polymer composite (mica/PS) by computer fitting of the dielectric results as:

(i) The plots for low mica content (<10 wt.%) composite do not exhibit distorted semicircle because the PS matrix is a dominant component whose dielectric constant is nearly independent of frequency (non-polar polymer).

(ii) The plot for 15 wt.% mica content composite showed a distorted semicircle shown in Figure (7). This semicircle is attributed to mica response to the applied frequency because mica is a polar mineral. The dielectric behavior of the composite may attributed to polarization produced by the dipole relaxation and interfacial space effects of mica particulates.

(iii) The plot for high mica content composite showed two extrapolated distorted semicircles shown in Figure (8) for 70 wt.% specimen. The semicircle plot in Figure (8a) is attributed to dipole polarization takes place at high frequency range, while the semicircle plot of Figure (8b) is attributed to the interfacial space charge polarization takes place at lower frequencies (1 kHz).

TABLE II The parameters calculated from fitting a semicircle

Specimens (*wt.*%)	$\tau \times 10^{-4}$ (*s*)	ε_s	ε_∞	β
15	2	3.96	3.31	0.97
70 (low freq.)	38	10.51	5.56	0.88
70 (high freq.)	0.29	7.131	3.87	0.76

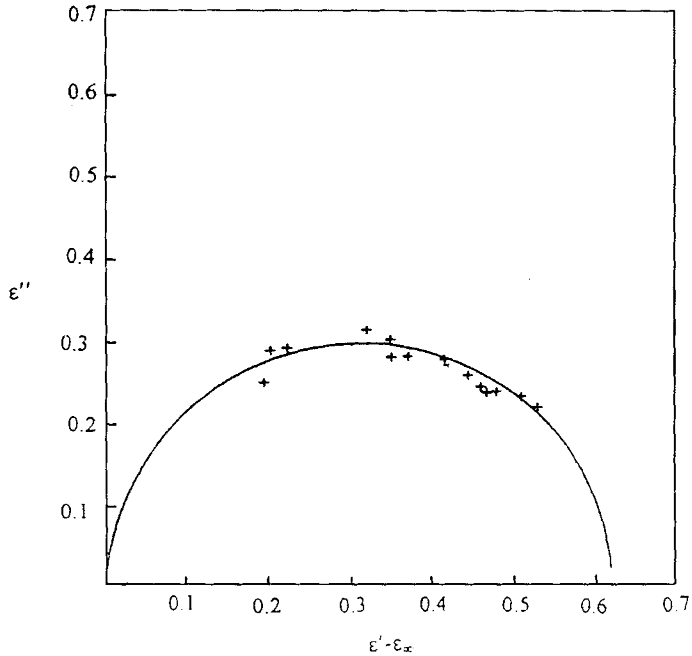

FIGURE 7 The Cole–Cole plot for the 15 wt.% specimen.

However, we can conclude from the Cole–Cole analysis that the observed relaxation effects and the polarization contribution arose from the dielectric behavior of the mica filler which is a polar material.

3.2. AC Conductivity *versus* Concentration

Few models were proposed to explain the electrical conductivity of polymer composites. Those models were based on statistical, geometrical and structure consideration. However, none of these models were fully successful in explaining all different results of the experimental studies performed on insulating or conductive polymer composites [20, 21].

An empirical relationship was proposed for describing the dependence of the electrical conductivity on volume fraction of the mica filler. The proposed relation is [21]

$$\sigma_{AC} = Ay^{\alpha} \tag{5}$$

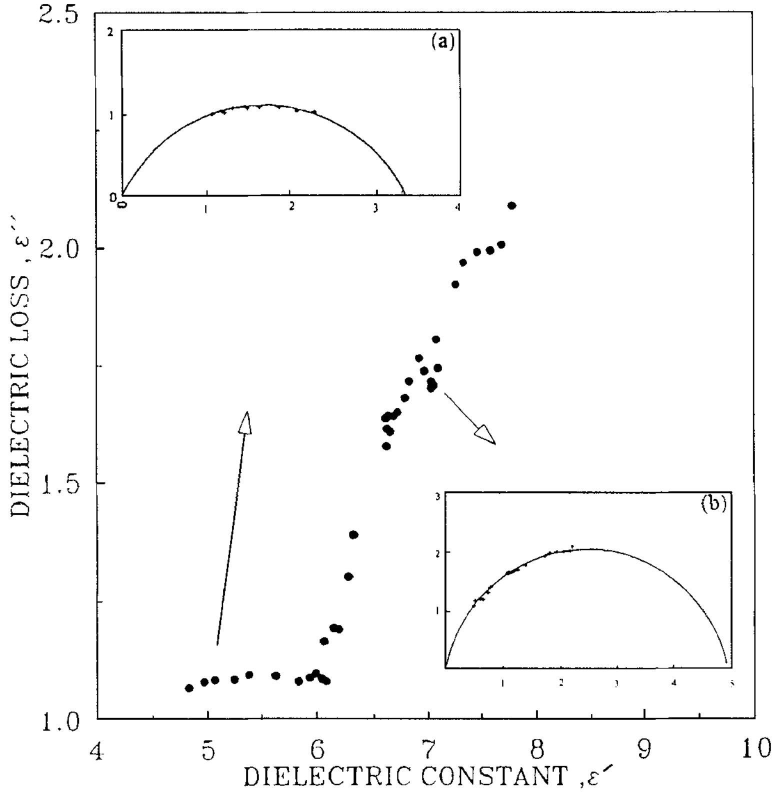

FIGURE 8 The Cole–Cole plot for the 70 wt.% specimen.

where y is the volume fraction of mica, α and A are constants which depend on the applied frequency. The volume fraction was calculated for the composite. Figure (9) shows a plot of the experimental data *versus* the volume content of mica. The solid line is obtained from the plot of Eq. (5) with the values of A and α as given in Table (III) which exhibits that both of them vary with the frequency of the applied field.

3.3. The Dielectric Constant *versus* Concentration

The dielectric constant, (ε^*) of particular composites can be calculated from the mixture formula involving the component of the dielectric constant, the volume fraction and the shape of the filler particles.

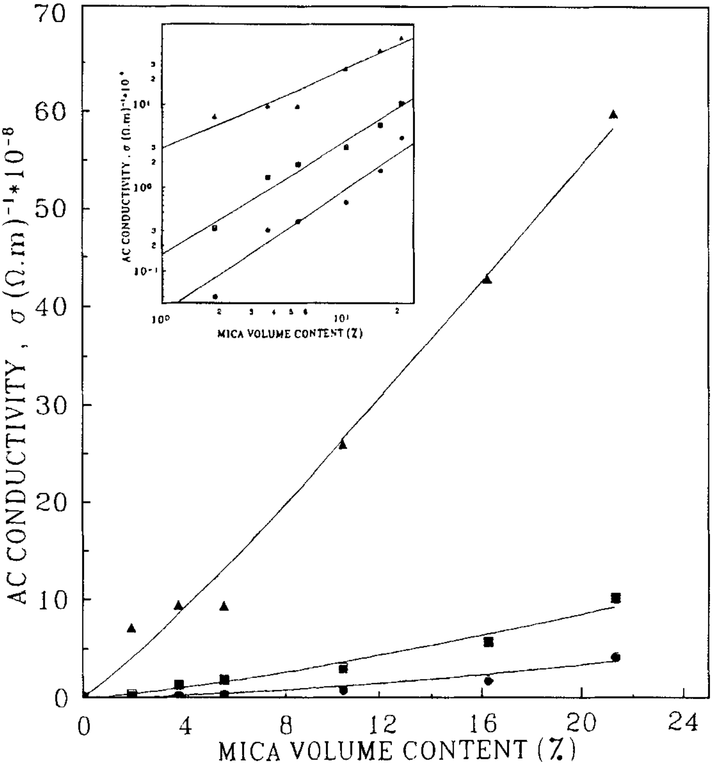

FIGURE 9 Fitting plot of the AC conductivity of the composites as a function of the volume content of mica at different frequencies.

TABLE III The estimated values of A and α

Frequency	$A \times 10^{-8} (\Omega \cdot m)^{-1}$	α
300 Hz	62.57	1.57
1 kHz	63.33	1.277
10 kHz	316.22	1.1

Thus, for a polymer matrix with dielectric constant, ε_1, incorporating uniformly distributed spherical inclusions of material with dielectric constant, ε_2, volume concentration of inclusion $(1-y)$, the dielectric constant of the composite can be estimated from the simplified Bruggesman's equation [14] which is an extension to Maxwell-Wagner formula [6, 22, 23].

$$\varepsilon^* = \varepsilon_1(1 + 3y) \tag{6}$$

Figure (10) shows that the above equation fits the observed dielectric constant rather well. The small difference between theory and experiments is due to that mica particles have a flake shape and not spherical as we assumed.

3.4. The AC Conductivity *versus* Frequency

Normally, analyzing the dependence of the observed AC conductivity can be done using the simplified formula [23–25]

$$\sigma_{AC} \approx Bf^n \tag{7}$$

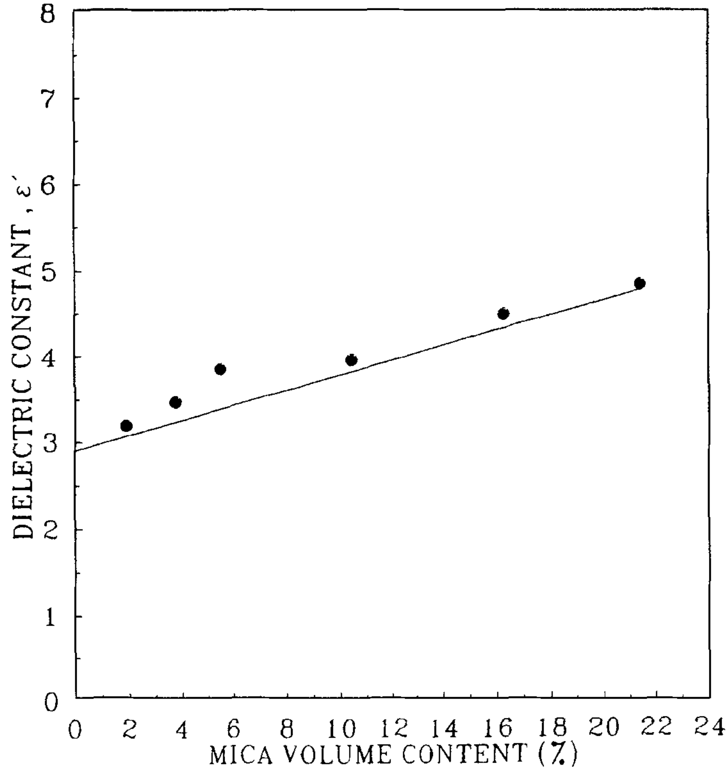

FIGURE 10 Fitting plot of the dielectric constant of the composites as a function of the volume content of mica at frequency 10 kHz.

where, n and B are conductivity coefficients. The fitting procedure is based on the least square method, and the estimated coefficients are listed in Table (IV). Figure (11) exhibits the fitting of Eq. (7) on the observed AC conductivity of the mica/PS composite as a function of frequency. This applicability may be an indication of that the conduction mechanism takes place in this polymer composite may be due

TABLE IV The estimated coefficient B and n

Specimens (*wt.*%)	$B \times 10^{-11} (\Omega \cdot m)^{-1} (Hz)^{-n}$	n
0.0	0.38	0.85
10	3.10	0.86
15	3.02	0.85
30	3.16	0.97
50	7.24	0.94
70	45.75	0.77

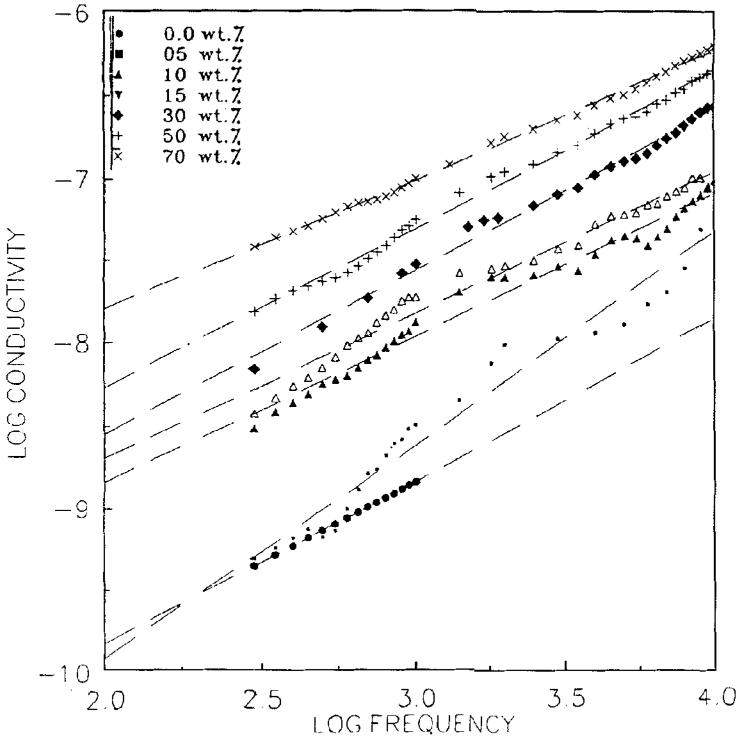

FIGURE 11 Fitting plot of $\log \sigma_{AC}$ *versus* $\log f$.

to hopping conduction between the extrinsic localized molecular-ion states which results from the existing impurities.

4. CONCLUSIONS

The research work presented in this paper deals with the electrical properties of mica/polystyrene composite. The electrical conductivity and the dielectric behavior were studied as a function of mica concentration and the applied electric field frequency. From the obtained results the following conclusion are:

1. The observed AC conductivity showed frequency and mica content dependence.
2. The dielectric constants showed mica content and frequency dependence.
3. The observed electrical results fit approximately the reported empirical equations concern the AC conductivity and dielectric behavior of polymer composites.
4. The observed relaxation and polarization effects of the composite are mainly attributed to the dielectric behavior of the mica filler.
5. The results were explained on the basis of the interfacial (space-charge) polarization, dipolar polarization and on the decreases of the hindrance of the polymer matrix.

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References

[1] Katz, H. S. and Milewski, J. V. (1978). "Handbook of Fillers and Reinforcements for Plastics", first edition, Van Nostrand Reinhold Company, New York, pp. 334–368.

[2] Goosey, M. T. (1985). "Plastic for Electronics", first edition, Elsevier Applied Science Publishers, London, pp. 1–49.

[3] Baker, Z. Q., Abdelazeez, M. and Zihlif, A. (1988). *J. Mater. Sci.*, **23**, 2995; Musameh, S. M., Ahmad, M. and Zihlif, A. (1992). *Mater. Sci. Eng.*, **B14**, 1–4.

[4] Long Wu, S. and Tung, I. (1995). *Polymer Comp.*, **16**, 233.

[5] Chaudry, M. A. and Jonscher, A. K. (1998). *J. Mater. Sci.*, **23**, 208.

[6] Banhegy, G., Rho, M., Chien, J. C. and Karasz, F. (1987). *J. Polym. Sci. Polym. Phys.*, **25**, 57.

[7] Hanly, D. P. and Aust, F. I. (1980). "Introduction to the Selection of Engineering Materials", first edition, Van Nostrand Reinhold Company, New York, pp. 73–110.
[8] Mijovk, J. and Bellucci, F. (1996). *Trends in Polymer Sci.*, **4**, 74–80.
[9] Ahmad, M., Zihlif, A., Martuscelli, A. and Ragosta, E. (1992). *Polym. Comp.*, **12**, 83–90.
[10] Hussain, A. and Zihlif, A. (1993). *J. Thermoplastic Comp. Mater.*, **6**, 120–129.
[11] Ahmad, A., Abdallah, M. and Zihlif, A. (1988). *Appl. Phys. Comm.*, **8**, 169–190.
[12] Shahin, M., Abdalla, M. and Zihlif, A. (1995). *J. Polym. Mater.*, **12**, 151.
[13] Tripathi, A. and Pilla, P. (1990). *J. Mater. Sci.*, **25**, 1947.
[14] Tareev, B. (1979). "Physics of Dielectric Material" first edition, Mir Publishers, Moscow, pp. 67–125.
[15] McCrum, N., Read, N. and Williams, G. (1967). "Anelastic and Dielectric Effects in Polymeric Solids", first edition, J. Wiley, London, pp. 102–140.
[16] Marton, L. and Marton, G. (1980). "Methods of Experimental Physics", Vol. 16, first edition, Academic press, London, pp. 379–395.
[17] Al-Ani, S., Al-Hassany, H. and Al-Dahan, Z. (1995). *J. Mater. Sci.*, **30**, 3720.
[18] Nayak, B., Mansingh, A. and Machwe, M. (1990). *J. Mater. Sci.*, **25**, 749.
[19] Palin, G. R., "Plastics for Engineering", first edition, 749 Chapman and Hall Ltd., London, pp. 1–97.
[20] Lux, F. (1993). *J. Mater. Sci.*, **28**, 285–301.
[21] Andretta, A., Heeger, A. and Smith, P. (1990). *Polym. Comm.*, **31**, 275.
[22] Hasted, J. B. (1975). "Aqueous Dielectric", first edition, Chapman and Hall, London, pp. 25–67.
[23] Othmer, K. (1982). "Encyclopedia of Chemical Technology", Vol. 18, third edition, J. Wiley, New York, pp. 755–793.
[24] Diaconu, I., Dumitrescu, S. and Simionescu, C. (1980). *Europ. polym. J.*, **16**, 511.
[25] Almond, D., Hunter, C. and West, A. (1984). *J. Mater-Sci.*, **19**, 3236.