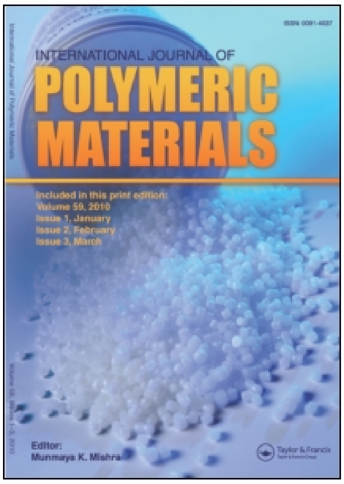

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A.C. Conductivity and Dielectric Properties of Polycarbonate Sheet

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A.C. Conductivity and Dielectric Properties of Polycarbonate Sheet

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The a.c. conductivity $\sigma_{a.c}(\omega)$, the dielectric constant ε' and dielectric loss ε'' of polycarbonate sheet have been studied in the frequency range 1.5×10^2–5×10^6 Hz and temperature range 294–473 K. A.c. conductivity $\sigma_{a.c}(\omega)$ results show that it depends strongly on the frequency and slightly on the temperature, with activation energy between 0.0003 and 0.085 e.V. The a.c. conductivity $\sigma_{a.c}(\omega)$ as a function of frequency is well described by a power law $A\omega^S$ where S is the frequency exponent. The obtained values of $S < 1$ in the first region over the frequency range 1.5×10^2–4×10^3 Hz, the temperature dependence of both a.c. conductivity $\sigma_{a.c}(\omega)$ and the parameter S are reasonably well interpreted by the correlated barrier hopping (CBH) model. The second region, where $S > 1$ over the frequency range 4×10^3–5×10^6 Hz, has a strong dependence on the frequency but near independence of the temperature, suggesting that the conductivity may be interpreted by the Maxwell–Wagner (M–W) dispersion. In the first region, the optical band gap of the material, E_g, and the maximum barrier height, W_m, are calculated, suggesting that there is agreement with that proposed by the theory of hopping of charge carriers over potential barrier as suggested. Spatial inhomogeneity may affect the conduction mechanisms in polycarbonate. The experimental values of dielectric constant, ε' and dielectric loss, ε'', show their dependence on temperature and frequency.

Keywords: polymer, polycarbonate, a.c. conductivity, dielectric properties

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INTRODUCTION

Polycarbonate is an example of a complex thermoplastic polymer, with a toughness, dimensionally stability and transparency properties, that has many applications in the last decades. Polycarbonate also has outstanding thermal stability, exceptional machinability, stain resistance, and non-toxicity with low water absorption. Polycarbonate is excellent for electrical applications because of its high dielectric strength and high volume resistivity that decrease only as the temperature is increased. The structure of polycarbonate is shown in Figure 1.

The magnitude of the dielectric constant ε' of polymers is determined by their chemical and physical structure and compositions. The parameters that characterize the dielectric loss, ε'', and loss tangent, tan δ, depend on the specific features of molecular motion of polymer and hence on their chemical and physical structure. In general, most of the polymers are insulators because of their low conductivity. The conductivity depends on thermally and electrically generated carriers and also on the addition of suitable dopants [1–2]. Generally, polymers are amorphous or semi-crystalline substances. In amorphous substances there are many localized trapping sites and the carrier mobility is very low. The transport mechanism in amorphous materials is more complicated than in crystalline materials, where a long range order exists [3].

The aim of this work is to study the a.c. conductivity, $\sigma_{a.c}(\omega)$, and dielectric constant, ε', and dielectric loss, ε'', to characterize the conduction mechanism of the polycarbonate sheet.

EXPERIMENTAL DETAILS

Material Used

Polycarbonate sheet was supplied by San Diego Plastics, Inc., Canada. The sample has a uniform thickness equal to 0.6 mm measured by

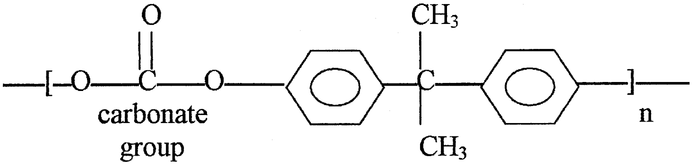

FIGURE 1 The structure of polycarbonate.

using sensitive micrometer. The polycarbonate sheet was cut into samples suitable for the required measurements.

A.C. Conductivity Measurements

The polycarbonate samples were covered on both sides with thin layer of a silver paste to obtain good contact and were mounted between two brass electrodes in a special holder. Then the sample holder was inserted in a small furnace and the temperature was measured to better than ±1°C with calibrated Ni–NiCr thermocouple that was set up near the sample. A.c. conductivity, dielectric constant and dielectric loss of the polycarbonate samples were calculated from the measured values of the impedance, Z, the capacitance C_p and the loss tangent, $\tan\delta$, using Hioki 3532 LCR meter.

The a.c. conductivity was calculated from the equation:

$$\sigma_{a.c}(\omega) = d/Za \tag{1}$$

where d is the sample thickness and a is the sample cross-section area.

The dielectric constant was calculated using the relationship:

$$\varepsilon' = C_p d/a\ \varepsilon_0 \tag{2}$$

where ε_0 is the free space permittivity.

The dielectric loss ε'' was calculated from the relationship:

$$\varepsilon'' = \varepsilon' \tan\delta \tag{3}$$

where $\delta = 90 - \Phi$, Φ is the phase angle that has been measured using the same LCR meter. The measurements were carried out in the frequency range 1.5×10^2–5×10^6 Hz in a controlled temperature range of 294–473 K.

RESULTS AND DISCUSSION

A.C. Conductivity

The frequency dependence of the a.c. conductivity $\sigma_{a.c}(\omega)$ of polycarbonate samples over the temperature range is shown in Figure 2. In this figure, two regions can be observed at all temperatures. The first region, measured at frequencies from 1.5×10^2 to 4×10^3 Hz exhibits temperature dependence and is nearly frequency insensitive due to onset of ionic conduction [4–5].

The second region, in frequencies exceeding 4×10^3 Hz, displays a strong frequency dependence and weak temperature dependence.

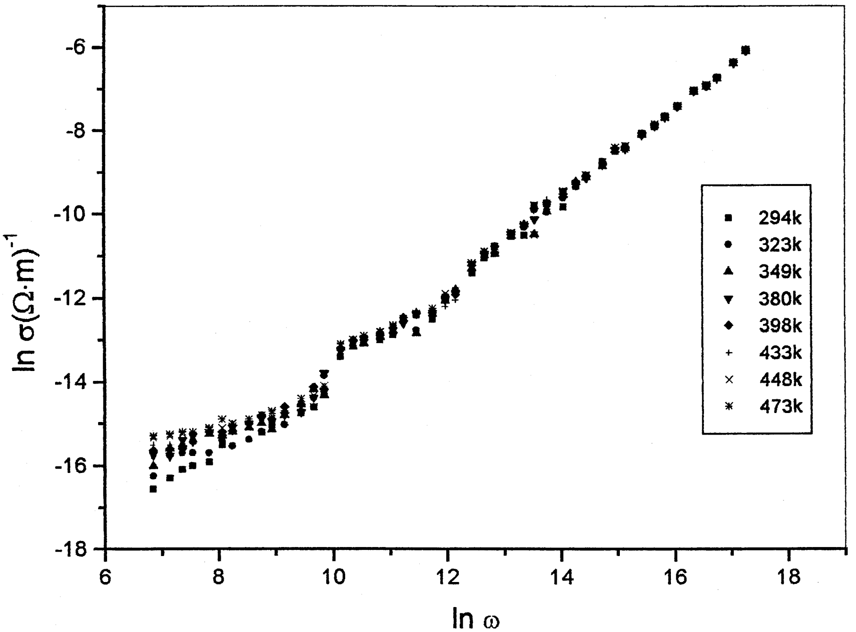

FIGURE 2 Frequency dependence of $\sigma_{ac}(\omega)$ for polycarbonate sheet at different temperatures.

The implication of both frequency and temperature dependence of a.c. conductivity is that the polymer sample follows the frequency dependence equation given by:

$$\sigma_{a.c}(\omega) = \sigma_0(T) + A(T)\omega^{S}(T) \qquad (4)$$

where $\sigma_0(T)$ is the d.c. conductivity contribution to $\sigma_{a.c}$ when the frequency f → 0, A(T) is a temperature-dependent constant, ω is the angular frequency, and S is the frequency exponent, and it is found to be less than unity in the first frequency range of 1.5×10^2–4×10^3 Hz, more than unity in the second frequency range of 4×10^3–5×10^6 Hz. Figure 3 shows the dependence of the exponent S on temperature in the first region. It can be observed that the values of S decrease with increasing temperature. The power S has the value of 0.71 at 294°C and reaches a value of 0.458 at 473°C. Therefore, the correlated barrier hopping model (CBH) is applicable to this sample [6–9]. According to the CBH model, values of S range from 0.7 to 1 at room temperature and are found to decrease with increasing temperature. This is in good agreement with the obtained results, so the

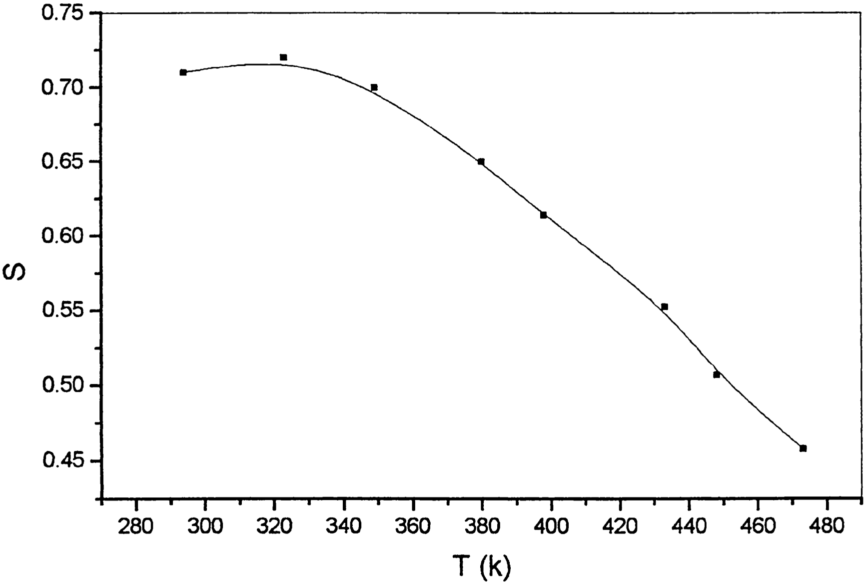

FIGURE 3 Temperature dependence of the frequency exponent S for polycarbonate sheet.

frequency dependent of $\sigma_{ac}(\omega)$ in the first region can be explained in terms of CBH model.

The expression for S derived on the basis of this model is given by [7–8,10].

$$S = 1 - 6\,kT/E_g \tag{5}$$

where k is the Boltzmann constant, T is the temperature in Kelvin, and E_g is the optical band gap of the material. From this approximation the values of E_g have been calculated and are given in Table 1.

In the second region $S > 1$ and nearly constant with temperature, suggesting that the conduction mechanism may be interpreted by the Maxwell–Wagner (M–W) dispersion [11]. The M–W dispersion is caused by interfacial polarization caused by the inhomogeneity of the material.

The temperature dependence of the conductivity $\sigma_{a.c}(\omega)$ at different frequencies for polycarbonate is shown in Figure 4. It is clear from the figure that $\sigma_{a.c}(\omega)$ increases linearly with the reciprocal of absolute temperature which follows the well-known equation:

$$\sigma = \sigma_0 \exp(-\Delta E_a/KT) \tag{6}$$

TABLE 1 Variation of the Maximum Barrier Height as a Function of Temperature in the Frequency Range 1.5×10^2–4×10^3 Hz

T (k)	E_g (e.V)
294	0.524
323	0.608
349	0.460
380	0.494
398	0.533
433	0.533
448	0.470
473	0.451

where σ_0 is a constant, ΔE_a the activation energy for conduction, K the Boltzmann's constant and T the absolute temperature. This suggest that the a.c. conductivity $\sigma_{a.c}(\omega)$ is a thermally activated process from different localized states in the gap or its tails [12]. The activation energy of conduction ΔE_a is calculated at different frequencies from

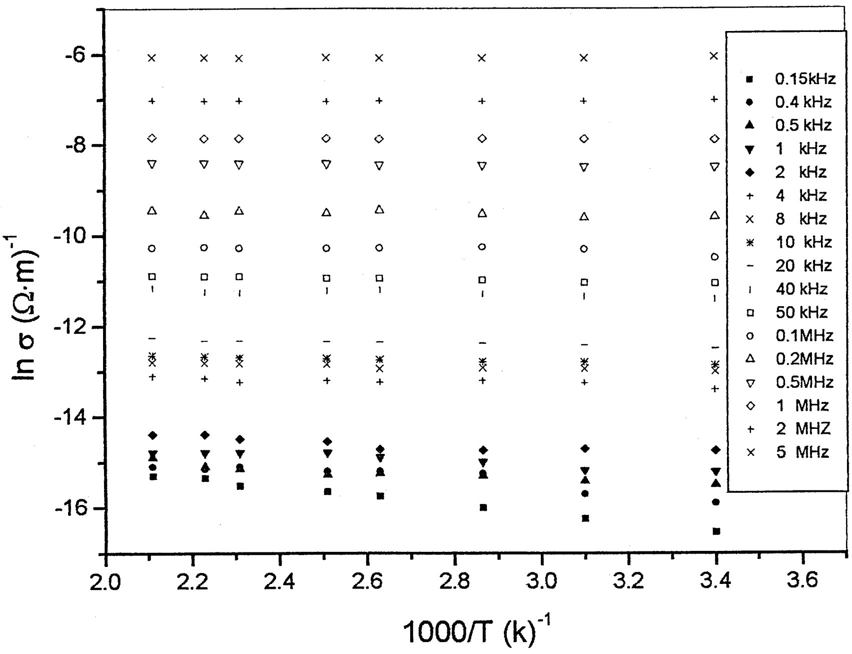

FIGURE 4 Temperature dependence of $\sigma_{ac}(\omega)$ for polycarbonate sheet at various frequencies.

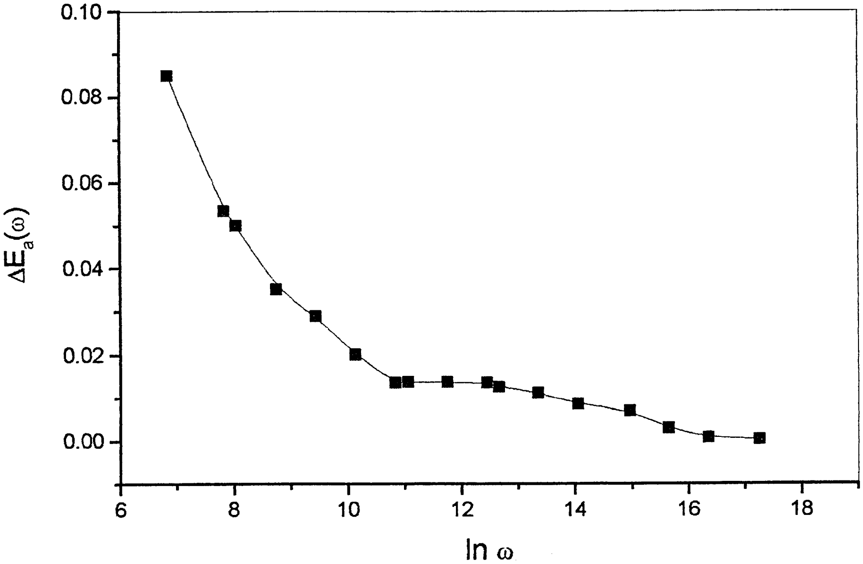

FIGURE 5 Frequency dependence of $\Delta E_a(\omega)$ for polycarbonate sheet.

the slopes of lines of Figure 4. The frequency dependence of the activation energy $\Delta E_a(\omega)$ for the investigated sample is shown in Figure 5. It can be seen that the activation energy $\Delta E_a(\omega)$ tends to decrease with increasing frequency as found for other amorphous materials [13]. The smaller values of the $\Delta E_a(\omega)$ and the increase of the a.c. conductivity with the increase of frequency confirm that hopping is the dominant mechanism.

Dielectric Studies

The dielectric constant ε' of polycarbonate sheets was measured as a function of frequency in the range 1.5×10^2–5×10^6 Hz, at different temperatures. In Figure 6, it can be noticed that ε' decreases with increasing frequency, verifying the fact that, for polar materials, the initial value of ε' is high but as the frequency of the a.c. field is raised, the value of ε'' begins to drop [14]. This decrease in the value of dielectric constant with frequency reaches a constant value at higher frequency corresponding only to interfacial polarization.

Further study on the relation between the dielectric constant and temperature is shown in Figure 7. This relationship is typical of polar dielectric materials where the orientation of the dipoles is facilitated

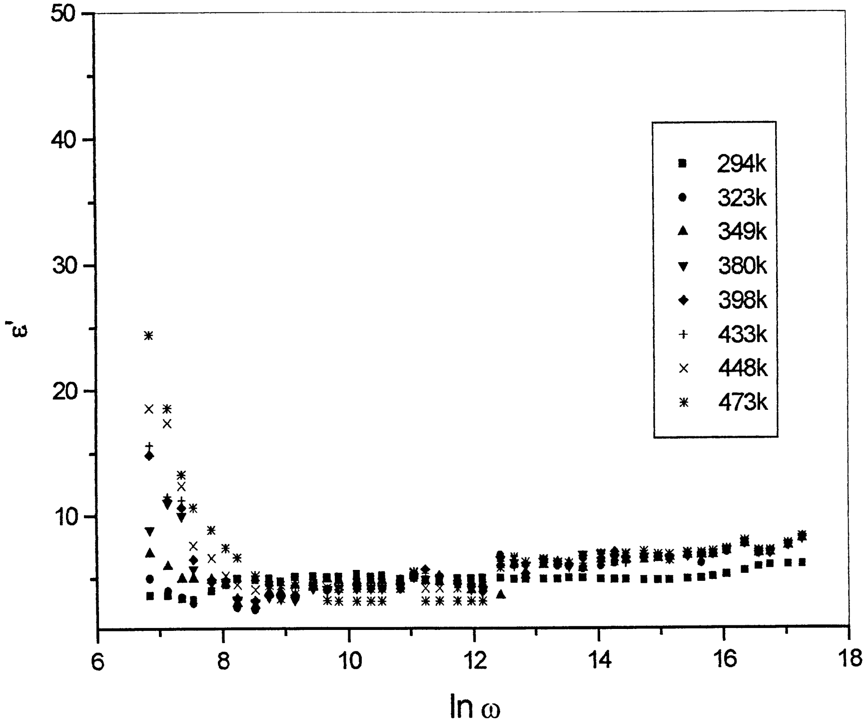

FIGURE 6 Frequency dependence of polycarbonate sheet at different temperatures.

by increasing temperature. The increase in ε' may also be caused by an increase in the degree of crystallinity [15–17], but not in the amorphous polycarbonate.

The occurrence of the shoulder of ε' at $T_p = 323\,K$ may be attributed to the decrease in the degree of ordering of polymer molecules due to chaotic thermal vibrations [18]. This shows that the relaxation frequency of the dispersion increases with increasing temperature and, as a response, moves to higher frequencies. This appears as an apparent increase in the permittivity at fixed frequency. The effect of increasing frequency on the dielectric constant is overruled by polarization and thus a decrease in the value of the dielectric of the system is observed. The dielectric constant increases with increasing temperature and the rate of increase is much faster at 5×10^6 Hz than at 2×10^6 Hz than at 1×10^6 Hz than at 1×10^5 Hz and 1×10^4 Hz [19]. This type of behavior may be due to disordering of the lattice, consisting of a shift of ions from one site to another.

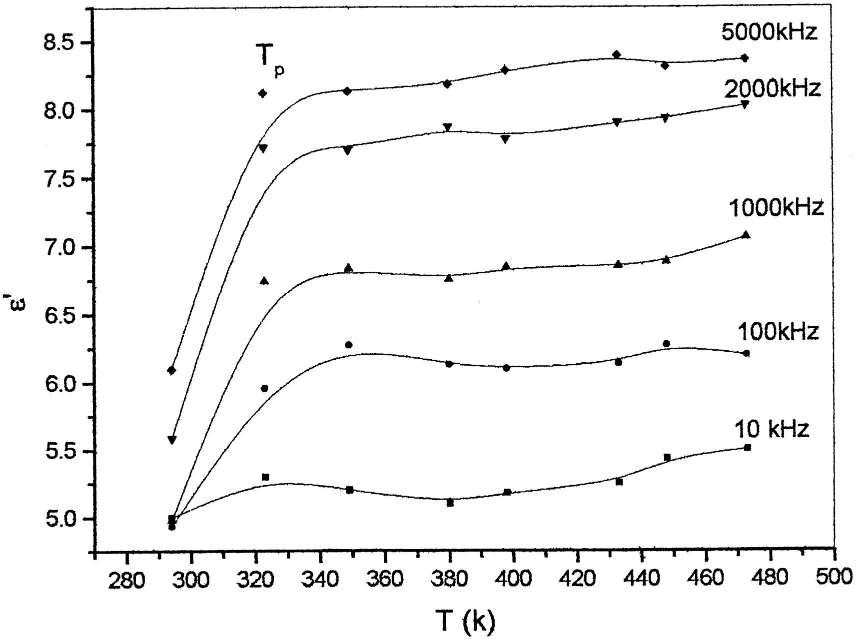

FIGURE 7 Variation of the dielectric constant with temperature of the polycarbonate.

The variation of the dielectric loss factor ε'' with the frequency at different temperatures is shown in Figure 8. The magnitude of the loss factor increases with increasing temperature. The increase of the loss factor at low frequency is due to dipole polarization. As the frequency increases the dipole polarization effect will tend to zero and then the dielectric loss factor tends to very small values, around 0.0001. On the other hand, ε'' decreases with increasing frequency following the formula:

$$\varepsilon'' = A\omega^{m} \tag{7}$$

where A is a constant and m is a power parameter that depends on the temperature. The power m is obtained from the negative slope of the straight lines of the relationship of ln ε'' versus ln ω as illustrated in Figure 9a. The variation of m with temperature is shown in Figure 9b. It is clear from this figure that m decreases linearly with temperature according to Giuntini's [20] equation:

$$m = -4\,kT/W_m \tag{8}$$

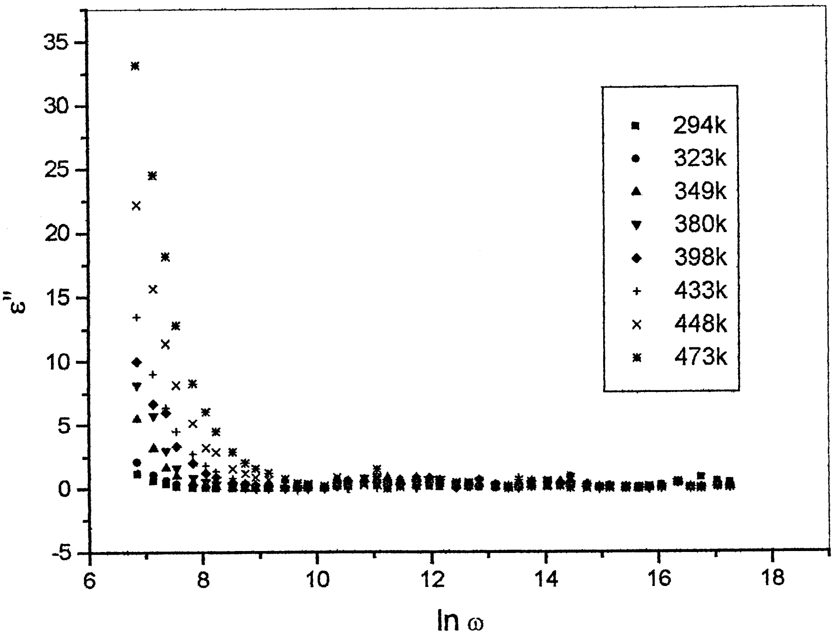

FIGURE 8 Frequency dependence of the dielectric loss polycarbonate sheet at different temperatures.

where W_m is the maximum barrier height. The obtained value of $W_m = (0.037\,\text{e.V})$ is in good agreement with the theory of charge carriers hopping over a potential barrier as suggested by Elliot [7–8]. The increase of ε'' with temperature can be also explained by Stavels [21] who divided the relaxation phenomenon into three parts: conduction losses, dipole losses, and vibrational losses. At low temperature, conduction losses have minimum value because it is proportional to (σ/ω). As the temperature increases, the a.c. conductivity $\sigma_{a.c}(\omega)$ increases and so the conduction losses increase, then the dielectric loss ε'' eventually increases.

CONCLUSION

Comprehensive studies of the a.c. conductivity $\sigma_{a.c}(\omega)$, the dielectric constant ε', and dielectric loss ε'' of polycarbonate sheet have been performed. The a.c. conductivity $\sigma_{a.c}(\omega)$ obeys a power law and can qualitatively be explained by the correlated barrier hopping model in the low frequencies and by Maxwell–Wagner dispersion in the high

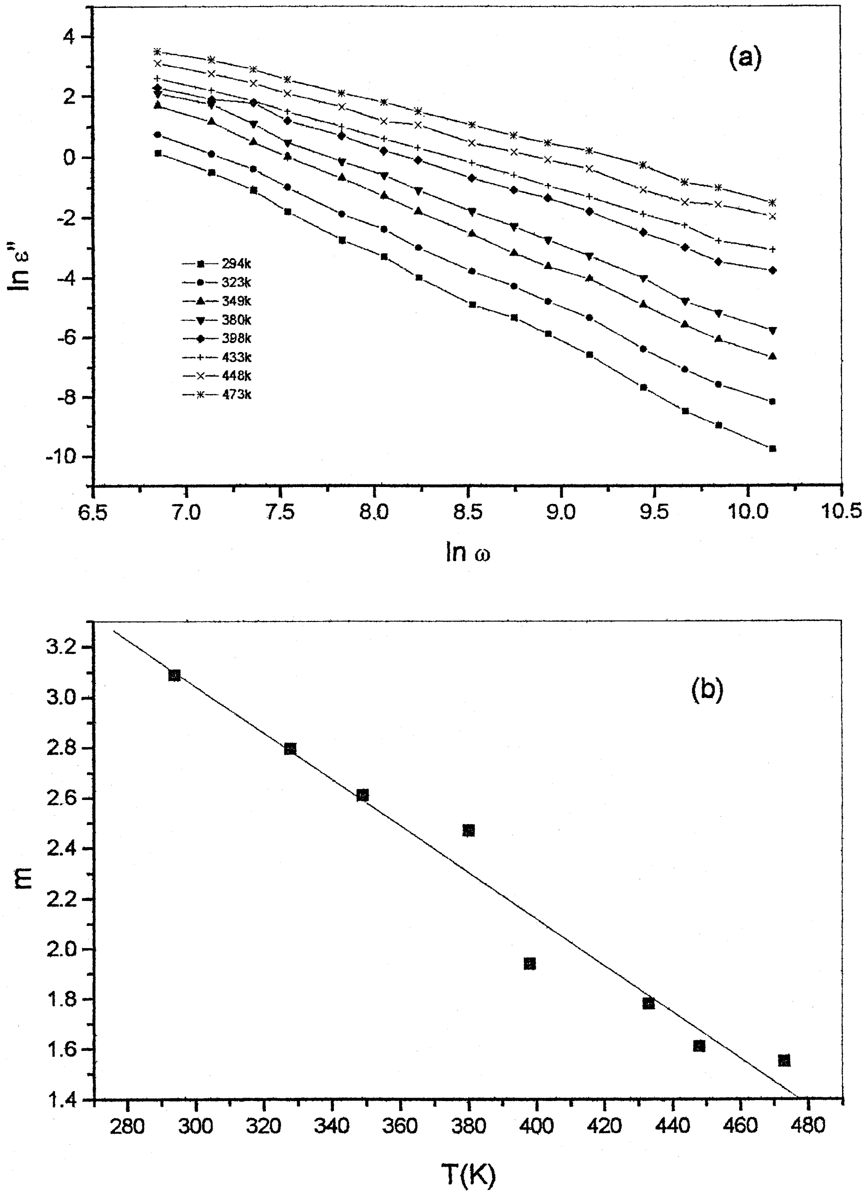

FIGURE 9 (a) ln ε'' versus ln ω for polycarbonate sheet at different temperatures. (b) The temperature dependence of the parameter.

frequencies. The dielectric constant ε' and dielectric loss ε'' of the sample decrease with increasing frequency and increase with increasing temperature. Thus, the dielectric constant ε' and the dielectric loss ε'' of the studied sample seem likely to be both frequency and temperature dependent. Frequency and temperature dependence of the dielectric constant ε' was attributed to interfacial and orientational polarization, respectively, whereas, the temperature dependence of the dielectric loss ε'' is associated with the conduction loss. The maximum barrier height is estimated from the data of dielectric loss, which is in good agreement with the theory of charge carriers hopping over a potential barrier between charged defect states.

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