

Dielectric Investigation of Some Normal Alcohols and Diols Dispersed in Some Polymeric Matrices

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ABSTRACT: A series of diols and normal alcohols were dispersed in some polymeric matrices, namely, polystyrene (PS), polyethylene (PE), and poly(methyl methacrylate) (PMMA). The dielectric properties of these systems were investigated in the frequency range of 10^2 – 10^5 Hz at a room temperature of 25°C. The experimental data were analyzed according to the Fröhlich equation into two relaxation processes: The first relaxation in the lower-frequency range could be attributed to the Maxwell–Wagner effect as a result of the difference in permittivities and conductivities of the investigated systems. The second relaxation in the higher-frequency range could be attributed to an intramolecular motion involving the rotation of various segments of the chain about the C–C bond accompanied by movement of the main dipoles for either diols or alcohols. The presence of the phenyl ring in the PS matrix may hinder such rotation when compared with the other matrices. The dielectric properties were also investigated for PMMA blended with either PS or PE at different ratios before and after the addition of small quantities of ethanediol. The interaction that might be expected between PMMA and PS in the blend may affect the rotation of the ethanediol chain in the blend matrix. The dielectric data as well as the data obtained from the calculated heat of mixing show that the investigated blends are incompatible. The addition of ethanediol to such blends gives the possibility that a large number of dipoles can be impregnated into the blend matrix and the problem of phase separation could be solved. © 2002 Wiley Periodicals, Inc. *J Appl Polym Sci* 85: 271–281, 2002

Key words: dielectric properties; relaxation; blends; compatibility

INTRODUCTION

The dielectric properties of alcohols and diols have attracted the attention of many workers. There is great agreement that these molecules associate into various polymeric species.¹ The

precise nature of these species and the parameters governing the various associative equilibria are still very much debatable.

Dielectric relaxation studies on liquid alcohols and diols have revealed that the dielectric dispersion is attributed to the OH group reorientation, whereas the intermediate dispersion is suggested to be a contribution from a molecular rotation due to an end group of a linear polymer and/or a smaller polymer reorientation. The low-frequency

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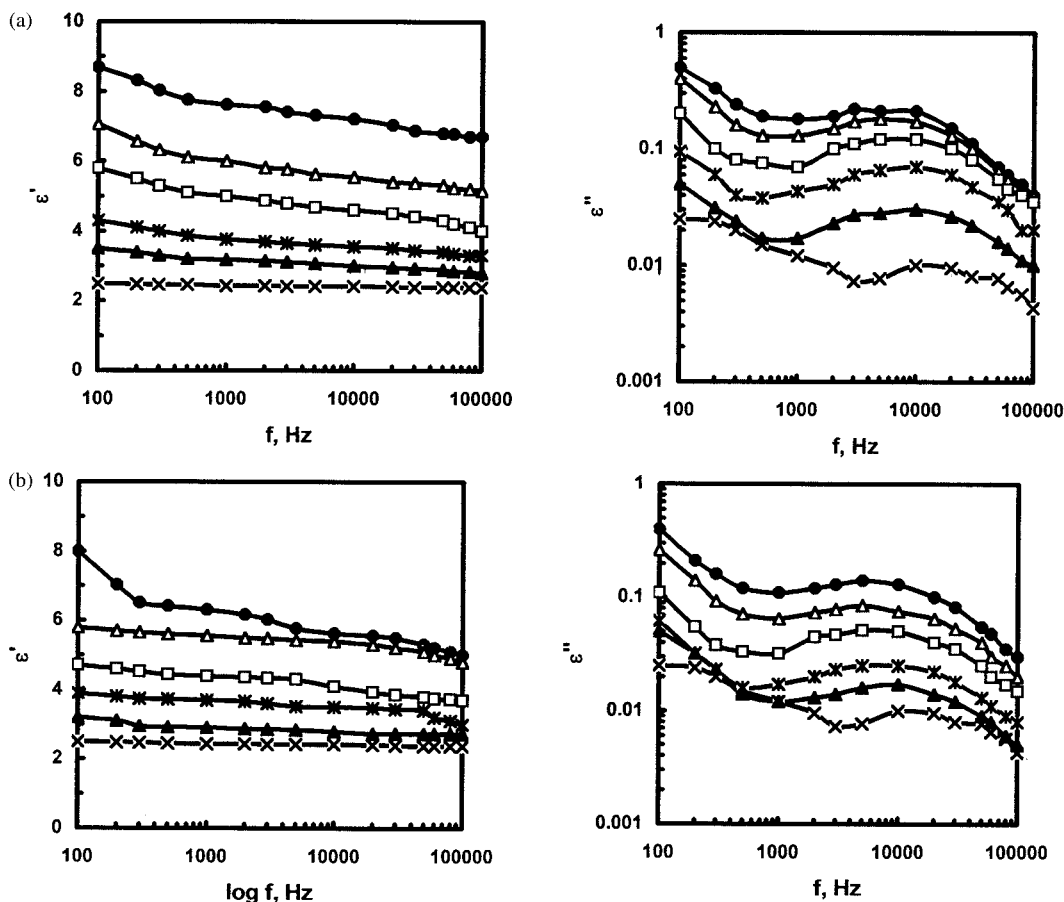


Figure 1 (a) Variation of permittivity ϵ' and dielectric loss ϵ'' with the applied frequency for (\times) PS and PS with 3% of different diols: (\bullet) ethanediol; (Δ) propanediol; (\square) butanediol; ($*$) hexanediol; (\blacktriangle) dodecanediol. (b) Variation of permittivity ϵ' and dielectric loss ϵ'' with the applied frequency for (\times) PS and PS with 3% of different alcohols: (\bullet) ethanol; (Δ) propanol; (\square) butanol; ($*$) hexanol; (\blacktriangle) dodecanol.

dispersion is a dominated process and is characterized by a simple Debye nature.²⁻⁸

Altogether, numerous studies have been carried out on various types of alcohols as pure liquid, in solution, and even in the solid state. However, very few works^{9,10} have been reported on alcohols in a matrix.

Intensive work has recently been carried out in the field of polymer blends.¹¹⁻¹⁸ This new investigating field of research has dealt with a great number of problems in the commercial explosion of polymer blending as an approach for the preparation of materials with newly described properties absent in the component polymers. Blending polymers is not an easy process due to the incompatibility of most of these blends, which reduce or extend the properties aimed from such blending.¹⁹ For this reason,

recent research work has been directed to improve the problem of phase separation of polymer blends.^{20,21}

The dispersion of alcohols at low concentrations in a matrix offers the possibility of separating the intramolecular forces from other processes.⁹ For this reason, we aimed to study the dielectric properties of polystyrene and polyethylene as nonpolar polymers and poly(methyl methacrylate) as a polar one before and after the addition of a small amount of different series of alcohols and diols to their matrices. It was also aimed to study the dielectric properties of the blends of poly(methyl methacrylate) with either polystyrene or polyethylene before and after the addition of a small quantity of ethanediol as a trial to improve the compatibility of such blends.

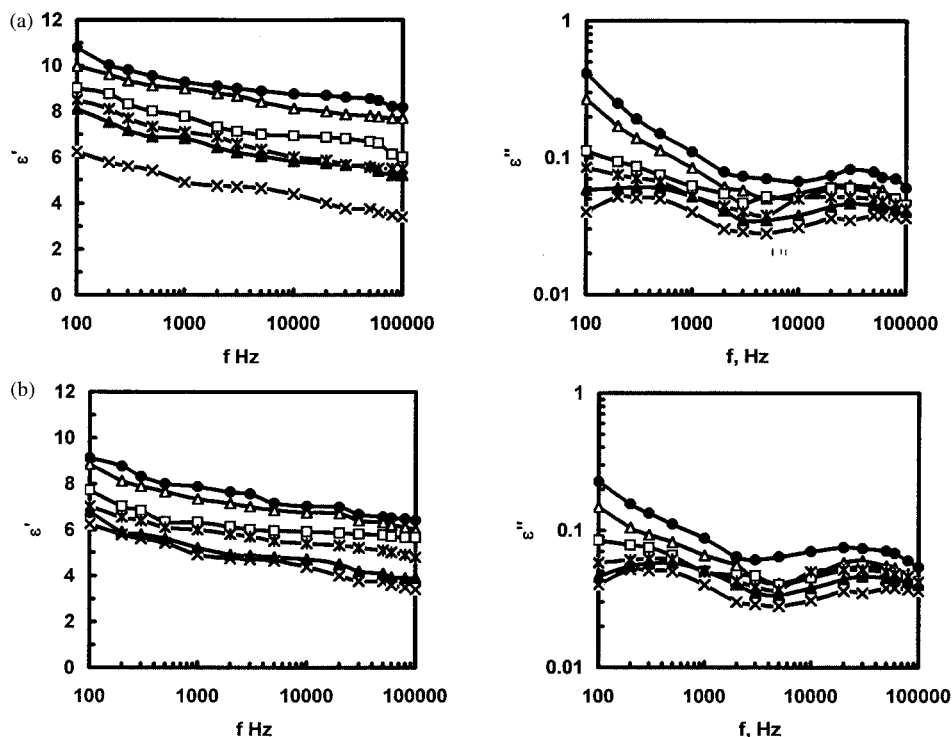


Figure 2 (a) Variation of permittivity ϵ' and dielectric loss ϵ'' with the applied frequency for (x) PE and PE with 3% of different diols: (●) ethanediol; (△) propanediol; (□) butanediol; (*) hexanediol; (▲) dodecanediol. (b) Variation of permittivity ϵ' and dielectric loss ϵ'' with the applied frequency for (x) PE and PE with 3% of different alcohols: (●) ethanol; (△) propanol; (□) butanol; (*) hexanol; (▲) dodecanol.

EXPERIMENTAL

Materials

Polystyrene (PS), low-density polyethylene (PE), and poly(methyl methacrylate) (PMMA) were obtained from Aldrich Chemical Ltd. (England). Ethanediol, propanediol, butanediol, hexanediol, octadecanediol, ethanol, propanol, butanol, hexanol, octadecanol, carbon tetrachloride, and chloroform were obtained from BDH (England). All chemicals were used as received.

Sample Preparation

A solution of LDPE was prepared by dissolving it in carbon tetrachloride and was divided into several fractions. The diols and alcohols were added in the same concentration of 3% by weight of the investigated polymer. Films were obtained by the casting method with a thickness of 0.17–0.22 mm.

PMMA and PS films were cast from chloroform. Blends were prepared by mixing the two

solutions with ratios of 75/25, 50/50, and 25/75 by volume to obtain clear films with thicknesses ranging from 0.17 to 0.22 mm.

Technique

Dielectric measurements were carried out in the frequency range of 100 Hz to 100 kHz using an LCR meter type AG-411 B (Ando Electric Ltd., Japan). The capacitance C , loss tangent $\tan \delta$, and ac resistance measured directly from the bridge from which the permittivity ϵ' , dielectric loss ϵ'' , and R_{dc} were calculated. The cell used was a parallel-plate circular condenser made of copper of an 8-mm diameter attached to a movable micrometer, similar to that designed by Fernandez.²² The cell was calibrated using standard materials (trolitul, glass, and air) with different thicknesses ranging from 1 to 5 mm. For each sample, a relation between the thickness d and its capacitance C_M was plotted as a standard curve. The capacitance C_M for the standard materials obtained from the standard curves was plotted versus the known permittivity ϵ' of each material

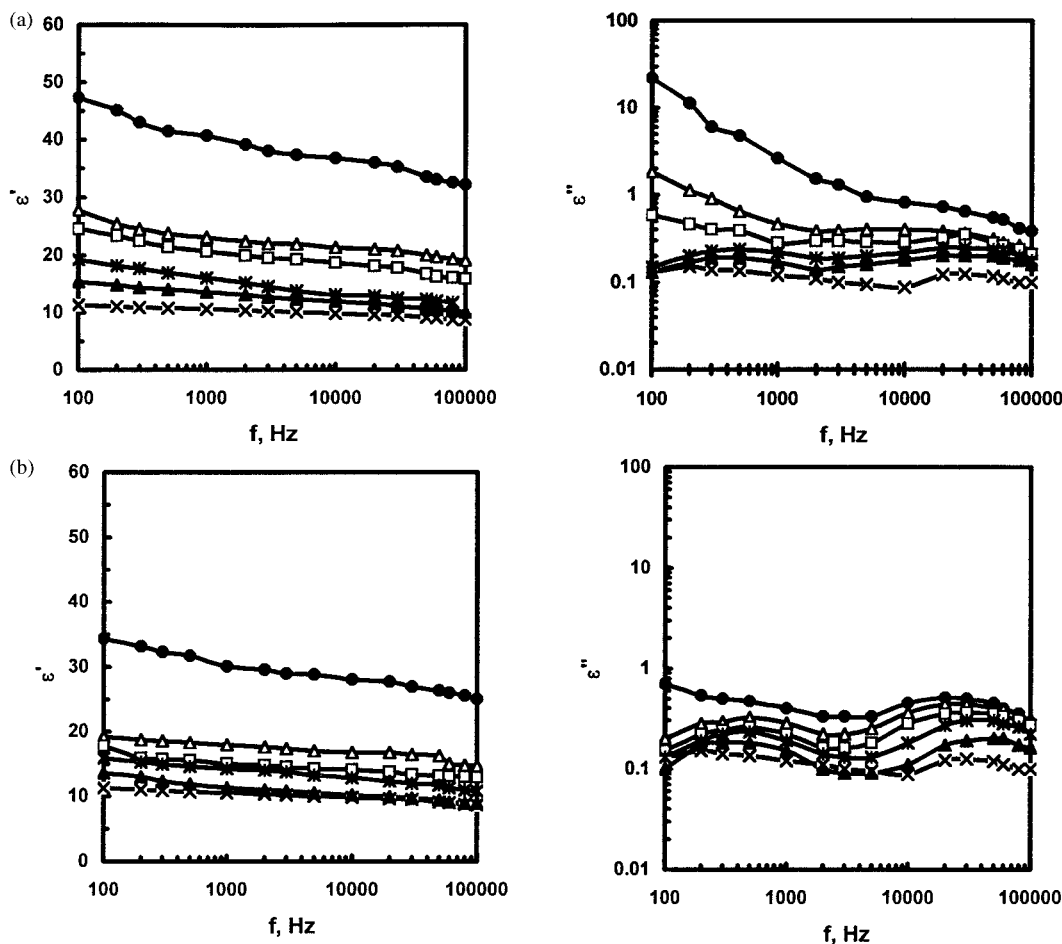


Figure 3 (a) Variation of permittivity ϵ' and dielectric loss ϵ'' with the applied frequency for (x) PMMA and PMMA with 3% of different diols: (●) ethanediol; (Δ) propanediol; (□) butanediol; (*) hexanediol; (▲) dodecanediol. (b) Variation of permittivity ϵ' and dielectric loss ϵ'' with the applied frequency for (x) PMMA and PMMA with 3% of different alcohols: (●) ethanol; (Δ) propanol; (□) butanol; (*) hexanol; (▲) dodecanol.

($\epsilon' = 2.5, 7, 1$ for trolitul, glass, and air, respectively). The relation between C_M and ϵ' was found to be linear and, thus, the permittivity corresponding to any measured capacitance can be deduced. To check the standard curve, two Teflon samples ($\epsilon' = 2.0$)²³ with different thicknesses were used. The experimental errors for ϵ' and ϵ'' were found to be ± 3 and ± 5 %, respectively.

RESULTS AND DISCUSSION

PS, PE, and PMMA Dispersed with Some Diols and Alcohols

The dielectric study was carried out for PS and PE as nonpolar polymers and PMMA as a polar

one before and after dispersing a series of diols and alcohols of small amounts (3% by weight) to their matrices. The added diols were ethanediol, propanediol, butanediol, hexanediol, and octadecanediol, while the added alcohols were ethanol, propanol, butanol, hexanol, and octadecanol.

The permittivity ϵ' and dielectric loss ϵ'' for the investigated polymers before and after the addition of diols and alcohols are plotted graphically in Figures 1–3 versus the applied frequency. The measurements were carried out at a room temperature of $\approx 25^\circ\text{C}$. From these figures, it is clear that the values of ϵ' decreased with an increase in the the applied frequency, showing anomalous dispersion. A pronounced increase in ϵ' is noticed by the addition of either ethanol or ethanediol to

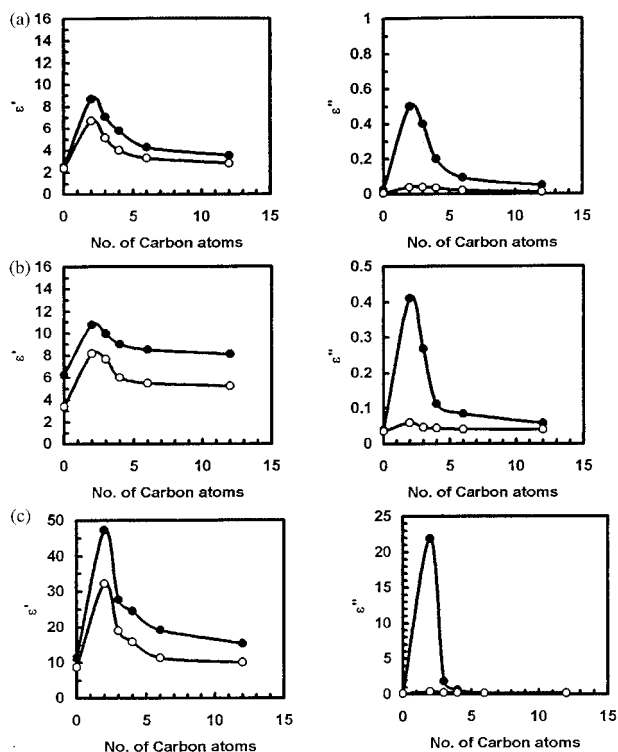


Figure 4 (a) Variation of permittivity ϵ' and dielectric loss ϵ'' with the number of carbon atoms in the added diols to PS: (○) $f = 100$ Hz; (●) $f = 100$ kHz. (b) Variation of permittivity ϵ' and dielectric loss ϵ'' with the number of carbon atoms in the added diols to PE: (○) $f = 100$ Hz; (●) $f = 100$ kHz. (c) Variation of permittivity ϵ' and dielectric loss ϵ'' with the number of carbon atoms in the added diols to PMMA: (○) $f = 100$ Hz; (●) $f = 100$ kHz.

the polymer matrix, which was found to decrease by increasing the number of carbon atoms in the added chain. This could be attributed to the number of dipoles per unit volume, which is expected to be large in the case of a small number of carbon atoms.^{24,25} This result is clearly detected when the values of ϵ' for different investigated matrices are plotted graphically versus the number of carbon atoms at two fixed frequencies of 100 Hz and 100 kHz as shown in Figures 4 and 5. From both figures, it is also noticed that the values of ϵ' for the diols are higher than are those for the alcohols.²⁶ This is an acceptable result as the values of the static permittivity of diols [ethanediol = 40.8, propanediol = 35.6, butanediol = 30.9, and hexanediol = 27.5 (ref. 26)] were found to be higher for pure alcohols [ethanol = 24.3, propanol = 20.1, butanol = 10.9, and hexanol = 8.97 (refs. 1 and 26)].

The absorption curves relating ϵ'' and the applied frequency shown in Figures 1–3 indicate that more than one relaxation mechanism is present. To the first approximation, analysis of the absorption curves was done in terms of the superposition of two Fröhlich terms according to the Fröhlich equation²⁷

$$\epsilon''(\omega) = \sum_{i=1}^n \frac{\epsilon_s - \epsilon_\infty}{P_i} \arctan \left[\frac{\sinh P_i/2}{\cosh \ln(\omega \bar{\tau}_i)} \right]$$

where P_i is a parameter describing the width of the distribution of relaxation times and equals $\ln(\tau_1/\tau_2)$. $\bar{\tau}_i$ is the mean relaxation time and equals $(\tau_1\tau_2)^{1/2}$. ϵ'' is the dielectric loss, while ϵ_s and ϵ_∞ are the static permittivity and the permittivity at infinite frequency, respectively.

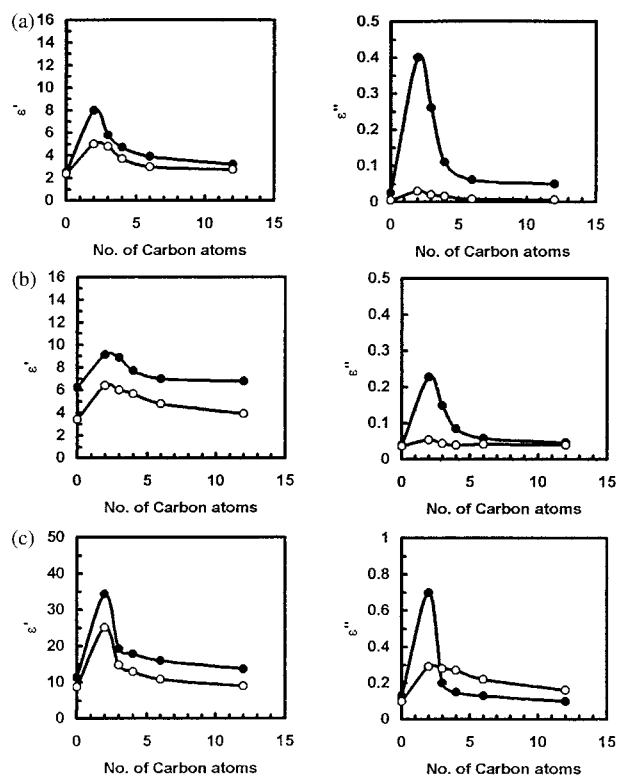


Figure 5 (a) Variation of permittivity ϵ' and dielectric loss ϵ'' with the number of carbon atoms in the added alcohols to PS: (○) $f = 100$ Hz; (●) $f = 100$ kHz. (b) Variation of permittivity ϵ' and dielectric loss ϵ'' with the number of carbon atoms in the added alcohols to PE: (○) $f = 100$ Hz; (●) $f = 100$ kHz. (c) Variation of permittivity ϵ' and dielectric loss ϵ'' with the number of carbon atoms in the added alcohols to PMMA: (○) $f = 100$ Hz; (●) $f = 100$ kHz.

Table I Conductivity and Relaxation Data of PS with Some Alcohols and Diols at Room Temperature

Added Molecule	$\sigma \times 10^{12}$ ohm ⁻¹ cm ⁻¹	ϵ''_m	$\tau_2 \times 10^6$ s
—	0.20	0.007	16.1
Ethanediol	3.33	0.200	32.1
Probanediol	2.20	0.170	27.2
Butanediol	1.00	0.120	25.0
Hexanediol	0.50	0.070	23.2
Octadecanediol	0.30	0.030	20.1
Ethanol	2.20	0.130	32.3
Propanol	1.04	0.080	27.2
Butanol	0.50	0.050	25.1
Hexanol	0.33	0.025	23.0
Octadecanol	0.28	0.017	20.0

For the data of the dc conductivity σ given in Tables I–III for the different investigated matrices, the dielectric loss ϵ''_σ at different frequencies ω was calculated using the equation²⁸

$$\epsilon''_\sigma = \frac{9 \times 10^{11} \times 4\pi\sigma}{\omega}$$

and subtracted from the experimental values of ϵ'' given at different frequencies. The data obtained revealed two distinct absorption regions: A low-frequency region was detected at a frequency lower than 500 Hz with the relaxation time τ_1 in the range 4×10^{-4} s. This region could be attributed to the Maxwell–Wagner effect,²⁹ which is expected to be at a lower-frequency range due to

Table II Conductivity and Relaxation Data of PE with Some Alcohols and Diols at Room Temperature

Added Molecule	$\sigma \times 10^{12}$ ohm ⁻¹ cm ⁻¹	ϵ''_m	$\tau_2 \times 10^6$ s
—	0.30	0.039	3.1
Ethanediol	20.00	0.080	5.3
Probanediol	12.50	0.060	5.0
Butanediol	4.00	0.055	4.5
Hexanediol	2.50	0.00	4.0
Octadecanediol	1.10	0.045	3.5
Ethanol	10.0	0.073	5.3
Propanol	6.10	0.055	5.0
Butanol	2.50	0.050	4.5
Hexanol	1.00	0.046	4.0
Octadecanol	0.50	0.042	3.5

Table III Conductivity and Relaxation Data of PMMA with Some Alcohols and Diols at Room Temperature

Added Molecule	$\sigma \times 10^{12}$ ohm ⁻¹ cm ⁻¹	ϵ''_m	$\tau_2 \times 10^6$ s
—	0.35	0.115	4.0
Ethanediol	1200.00	0.600	8.8
Probanediol	87.00	0.330	6.9
Butanediol	21.00	0.300	5.9
Hexanediol	10.00	0.250	5.0
Octadecanediol	5.00	0.200	4.8
Ethanol	27.00	0.500	6.4
Propanol	0.90	0.430	5.3
Butanol	0.60	0.350	4.8
Hexanol	0.40	0.300	4.5
Octadecanol	0.30	0.200	4.2

the multiconstituents of the investigated systems. The difference in permittivities and conductivities in the constituents of the polymeric materials is considered to be the reason for the presence of such an effect. On the other hand, the presence of impurities such as catalysts, antioxidants, and others that are usually added to the polymers through the polymerization process³⁰ could be the reason for this finding.

The second region in the higher-frequency range could be attributed to an intramolecular motion that involves the rotation of various segments of the chain about the C–C bond, accompanied by movement of the main dipole for either diols or alcohols. This mechanism is comparable with that found before in the case of alkanols and 1-thiols⁹ and some other series of sterically hindered alcohols in the PS matrix.¹⁰ The data obtained from the analyses are given in Tables I–III. An example of the analyses is shown graphically in Figure 6 for PMMA, PMMA dispersed with 3% hexanediol, and PMMA dispersed with 3% hexanol. The relaxation time τ_2 given in the case of PS is found to be higher than that for PE and PMMA. The steric hindrance due to the presence of a phenyl ring in the side group of PS may be considered to be the reason for such an increase, that is, the molecular volume increases and, consequently, the relaxation time.

It is also noticed that the relaxation time τ_2 and the maximum loss ϵ''_m were found to be higher in the case of ethanediol and ethanol and gradually decreased by increasing the number of carbon atoms in the chain. This trend could be attributed to the decrease in the number of dipoles in the

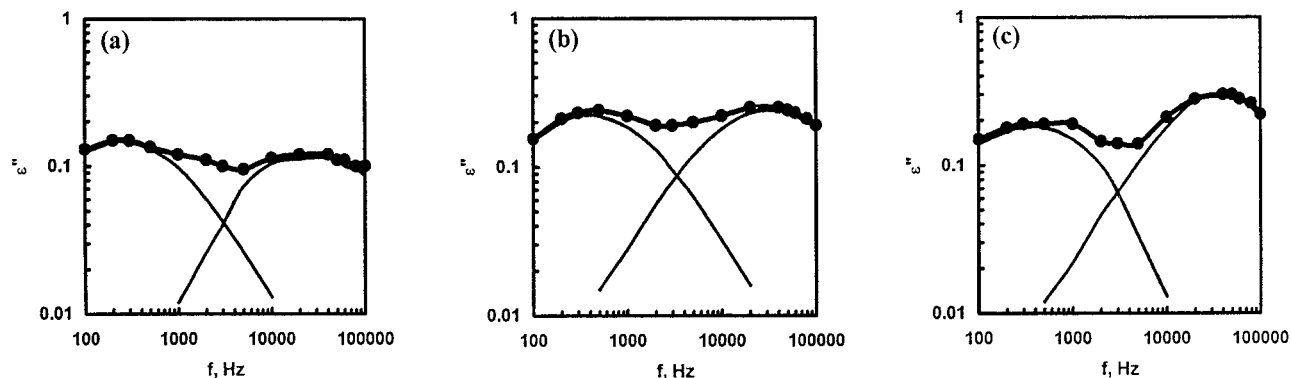


Figure 6 Absorption curves of (a) PMMA, (b) PMMA with 3% hexanediol, and (c) PMMA with 3% hexanol. (●) Fit of the experimental data using two Fröhlich terms.

matrix, which may lead to a decrease in the molar volume of the rotated molecule and, consequently, the relaxation time.^{31–33}

PMMA Blended with PS and PE Dispersed with Ethanediol

The permittivity ϵ' and dielectric loss ϵ'' obtained for PMMA blended with either PS or PE with

different ratios of 0/100, 25/75, 50/50, 75/25, and 100/0 are illustrated graphically in Figures 7 and 8 versus the applied frequency at room temperature. From both figures, it is clear that the values of ϵ' decrease by increasing either the applied frequency or the percentage of PS or PE in the blend.

The absorption curves relating ϵ'' and the applied frequency were analyzed using two Fröhlich

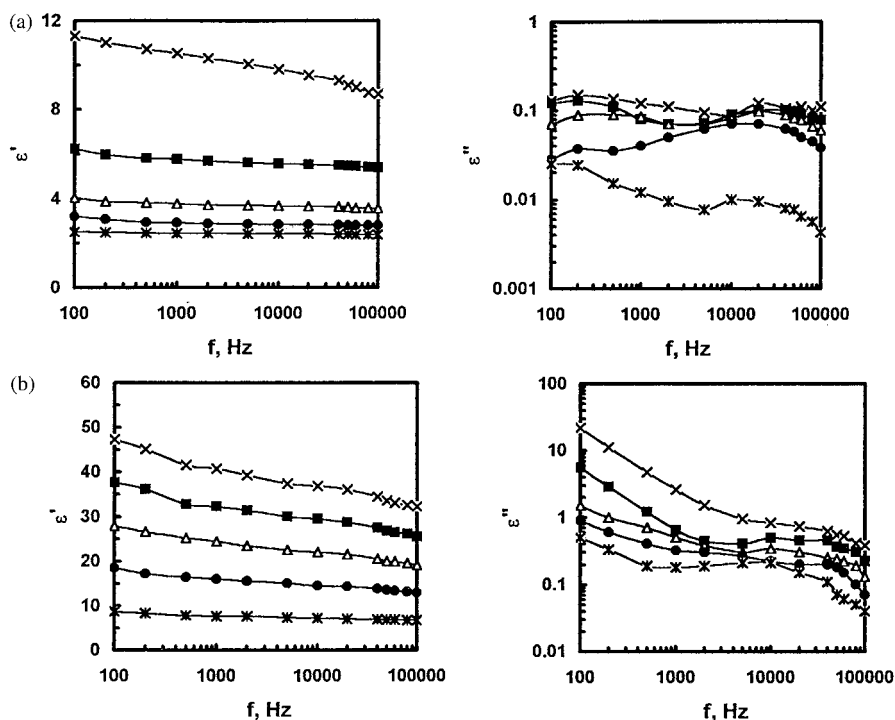


Figure 7 (a) Permittivity ϵ' and dielectric loss ϵ'' versus the applied frequency for PMMA/PS blends: (*) 0/100; (●) 75/25; (Δ) 50/50; (■) 25/75; (×) 100/0. (b) Permittivity ϵ' and dielectric loss ϵ'' versus the applied frequency for PMMA/PS blends with 3% ethanediol: (*) 0/100; (●) 75/25; (Δ) 50/50; (■) 25/75; (×) 100/0.

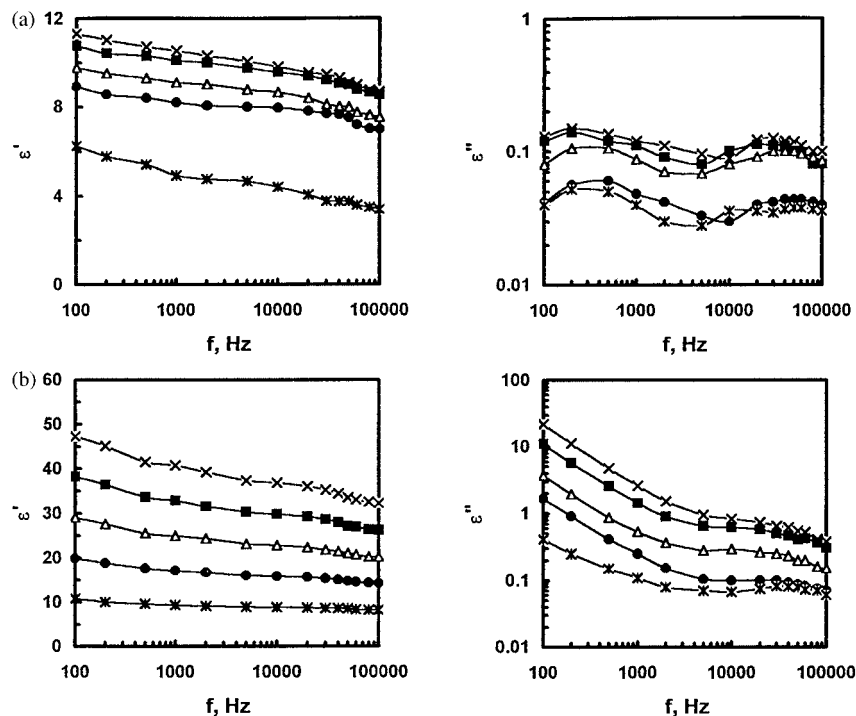


Figure 8 (a) Permittivity ϵ' and dielectric loss ϵ'' versus the applied frequency for PMMA/PE blends: (*) 0/100; (●) 75/25; (Δ) 50/50; (■) 25/75; (\times) 100/0. (b) Permittivity ϵ' and dielectric loss ϵ'' versus the applied frequency for PMMA/PE blends with 3% ethanediol: (*) 0/100; (●) 75/25; (Δ) 50/50; (■) 25/75; (\times) 100/0.

terms. The first term in the lower-frequency range could be related to the Maxwell–Wagner effect due to the difference in permittivities and conductivities of the investigated blend. The relaxation time τ_1 associated with that region lies in the order of $4 \epsilon' 10^{-4}$ s.

The second term detected at the higher-frequency range could be attributed to the side-group rotation since the main-chain motion is frozen at temperatures below the glass transition temperature. This finding is comparable with that found before by Takaaki and coworkers.³⁴ The data obtained from the analyses are given in Tables IV and V.

From both tables, it is clear that the relaxation time associated with such a region decreases by increasing the PMMA content in the PMMA/PS blend, while it is found to increase in the case of the PMMA/PE blend. The contrary result found here could be attributed to the presence of an interaction between the phenyl ring in PS with the ester group in PMMA. This may create a bigger free volume increase due to an increasing PS content in the blend.

To test the compatibility of those blends, the permittivity ϵ' is plotted graphically in Figure 9

versus the PMMA content in the blend at a frequency of 1 kHz. From this figure, it is clear that the values of ϵ' for the different ratios of the blends do not coincide on the line connecting the

Table IV Conductivity and Relaxation Data of PMMA/PS Blends With 3% Ethanediol at Room Temperature

PMMA Content	$\sigma \times 10^{12}$ $\text{ohm}^{-1} \text{cm}^{-1}$	ϵ''_m	$\tau_2 \times 10^6$ s
PMMA/PS			
100	0.35	0.115	4.0
75	0.30	0.100	5.3
50	0.28	0.090	6.5
25	0.22	0.070	8.8
0	0.20	0.009	16.1
PMMA/PS with 3% Ethanediol			
100	40.00	0.600	8.0
75	30.00	0.100	5.3
50	48.30	0.090	6.5
25	15.50	0.080	8.8
0	3.33	0.020	32.1

Table V Conductivity and Relaxation Data of PMMA/PE Blends With 3% Ethanediol at Room Temperature

PMMA Content	$\sigma \times 10^{12}$ ohm ⁻¹ cm ⁻¹	ε''_m	$\tau_2 \times 10^6$ s
PMMA/LDPE			
100	0.30	0.150	4.0
75	0.27	0.140	5.3
50	0.25	0.110	4.4
25	0.21	0.060	4.0
0	0.20	0.054	3.1
PMMA/LDPE with 3% Ethanediol			
100	1200.00	0.600	8.0
75	600.00	0.500	7.5
50	200.00	0.250	7.0
25	90.00	0.090	6.3
0	20.00	0.081	5.3

values of ε' of PMMA with either PS or PE. This nonlinearity indicates that the PMMA/PS and PMMA/PE blends are incompatible.³⁵⁻³⁷

The calculation of the heat of mixing is considered to be a tool for the determination of the degree of compatibility between polymer blends. It was noted that polymer compatibility in the solid state might occur if the heat of mixing is below 4.185×10^{-3} J/mol.³⁸ The heat of mixing was calculated for the systems under investigation using the following equation³⁸:

$$\Delta H_m = \{X_1 M_1 \rho_1 (\delta_1 - \delta_2)^2 [X_2 / (1 - X_2) M_2 \rho_2 + (1 - X_1) M_1 \rho_1^2]\}^{1/2}$$

where X , ρ , and M are the weight fraction of the polymer, the density, and the monomer unit molecular weight, respectively; δ is the solubility parameter of the polymers. The obtained values are illustrated graphically in Figure 10. It is clear from that figure that both the PMMA/PS and PMMA/LDPE blends are expected to be thermodynamically incompatible as the calculated heat of mixing lies above the upper limit of compatibility (41.8×10^{-3} J/mol). This result is considered to be good justification for the data obtained by the dielectric method.

Ethanediol was added at the same quantity (3% by weight) to the different compositions of the investigated blends. The permittivity ε' and dielectric loss ε'' were measured for such blends and

the data obtained are illustrated graphically in Figures 7 and 8 versus the applied frequency. ε' and ε'' are found to increase by increasing the PMMA content in the blend. By plotting ε' versus the PMMA content in both investigated blends at 1 kHz after the addition of ethanediol, as shown in Figure 9, it is interesting to find that the values of ε' merely coincide with the line connecting the values of ε' for the two individuals. This means that this addition could improve, to some extent, the compatibility of the investigated blends as its low number of carbon atoms gives the possibility of a large number of dipoles to be impregnated in the blend matrix. So, it could be concluded that ethanediol plays a big role in solving the problem of phase separation between the constituents of the blends and acts as a successful compatibilizer for such blends.

The higher values of ε'' noticed at the lower frequencies after the addition of ethanediol could be attributed to the presence of some sort of losses due to the dc conductivities. The values of ε''_{dc} for such blends were calculated from the data of conductivity σ given in Tables IV and V using the equation given above. The data were analyzed using the Fröhlich equation into two relaxation mechanisms: The first relaxation in the lower-frequency range, which is found to be in the order of 4×10^{-4} s, could be attributed to the Maxwell-Wagner effect. The second relaxation in the high-

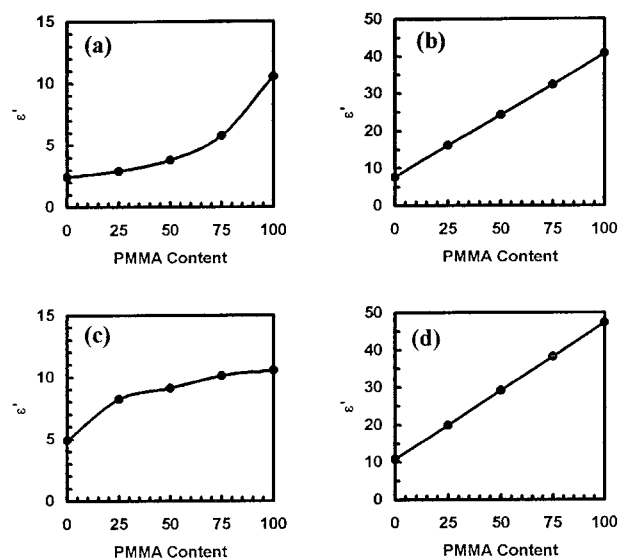


Figure 9 Relation between the permittivity ε' at 1 kHz and PMMA content in the blend: (a) PMMA/PS; (b) PMMA/PS with 3% ethanediol; (c) PMMA/PE; (d) PMMA/PE with 3% ethanediol.

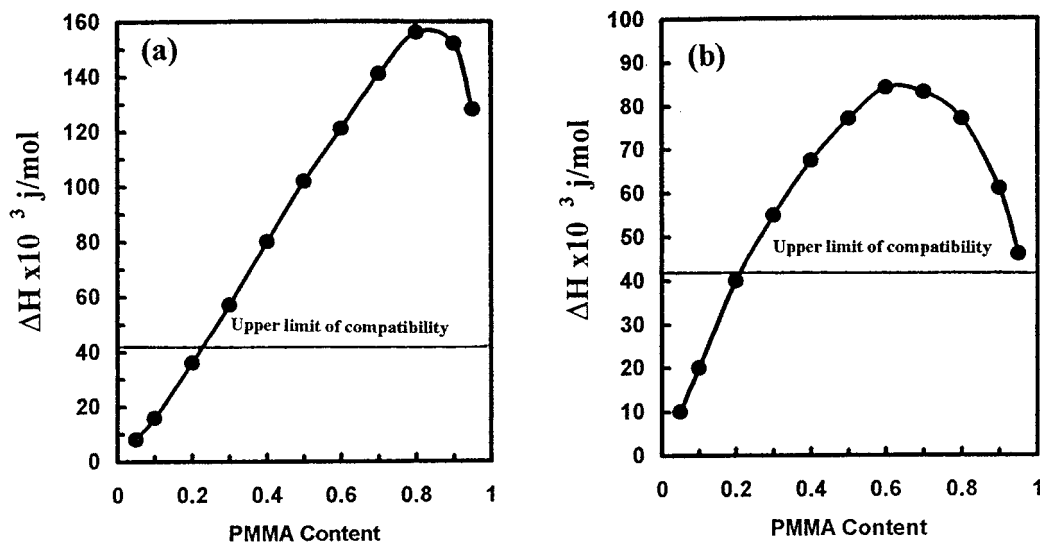


Figure 10 Relation between heat of mixing and PMMA content in the blend for (a) PMMA/PS and (b) PMMA/PE blends.

er-frequency range could be attributed to the rotation of various segments of the ethanediol chain about the blend matrix. The data obtained from the analyses are given in Tables IV and V. An example of the analyses is shown graphically in Figure 11 for the 50/50 PMMA/PS and PMMA/PE blends. The complex, which is expected to be formed between PMMA and PS, could give the alcoholic chain more chance to rotate in the blend matrix. This finding could explain the decrease in the relaxation time τ_2 from 32.1×10^{-6} to 8.8×10^{-6} s given in Table IV. This explanation can

be further supported by comparing the data with that of the PMMA/PS blend given in Table V where no pronounced change in the relaxation time τ_2 has occurred as no interaction is expected between the two polymeric materials.

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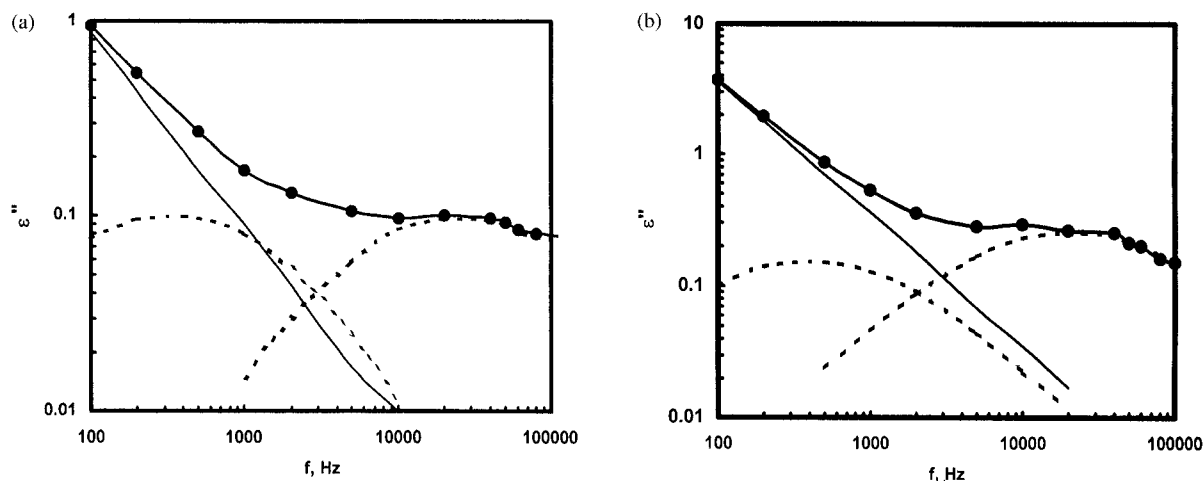


Figure 11 Absorption curves of (a) PMMA/PS (50/50) with 3% ethanediol and (b) PMMA/PE (50/50) with 3% ethanediol. Fit of the experimental data (●) using two Fröhlich terms (straight line) after subtraction of the dc losses.

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