

Electrical and Mechanical Properties of Some Polymeric Composites

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ABSTRACT: Two unsaturated polyester resins based on poly[propylene-phthalate-hexane-maleate] (PE1) and poly[oxydiethylene-phthalate-hexane-maleate] (PE2) were prepared and crosslinked with styrene monomer. The structure of the prepared polyesters was detected using IR and NMR. The thermal behavior of the styrenated polyesters was determined using differential scanning calorimetry. The dielectric properties for the PE1 and PE2 styrenated polyesters and their mixtures with different ratios were also studied with a frequency range of 100 Hz to 100 kHz at room temperature ($\approx 25^\circ\text{C}$). The mixture containing a 50/50 ratio of PE1/PE2 possessed the most promising dielectric properties. Thus, this sample was chosen along with the two separate styrenated polyesters to be loaded with three different types of fillers: calcium carbonate, clay, and quartz. This investigation led to the conclusion that the sample containing 50/50 PE1/PE2 loaded with 60–70% clay possessed the most promising dielectric properties. The compressive and tensile strength values were also studied for PE1, PE2, and their 50/50 mixture filled with the three types of fillers with the recommended concentrations (60 and 70%). The results indicated that the quartz composite (60%) had the best mechanical properties with respect to the clay and calcium carbonate. © 2002 John Wiley & Sons, Inc. *J Appl Polym Sci* 83: 1167–1180, 2002

Key words: electrical properties; mechanical properties; polymeric composites; fillers

INTRODUCTION

Polyesters are becoming increasingly popular because of their low cost, ease of use, and versatility. Some of their applications are in laminates, castings, art objects, industrial construction, insulation, embodiments, molding compounds, coatings, and adhesives.¹ Other advantages of polyesters are their high impact resistance and good weathering resistance.

Fillers are used with polyesters to reduce their cost and to improve some of their properties such as modifying the processing properties; increasing the hardness, thermal conductivity, and wear and chemical resistance; decreasing the thermal expansion; and improving the insulation properties. Various fillers such as quartz, calcium carbonate, and clays can be used without affecting the crosslinking performance.^{2,3}

The use of alumina trihydrate (ATH) or silica (SiO_2) as a filler in an unsaturated polyester resin reduces the flammability and enhances the electrical characteristics of the composites as compared to the styrenated polyester alone.⁴

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Table I Specifications for Clay, Calcium Carbonate, and Quartz

Clay (%)		Calcium Carbonate (%)		Quartz (%)	
Loss on ignition	9–11	CaO	55–56	Silica	99.4
SiO ₂	47–49	CO ₂	43–44		
Al ₂ O ₃	29–32	Fe ₂ O ₃	0.01–0.02		
Fe ₂ O ₃	4–8				
CaO	11.5				
MgO	0.2–0.9				

Hirano⁵ reported on an impregnated polymer in porous ceramics of fired clay tile or brick. The mechanical strength of this product at elevated temperatures was twice as high as that of unimpregnated material.

The effect of radiation-initiated curing of the polyester on the physicomechanical properties of a sand/clay-polyester composite was investigated by Ismail et al.⁶ They found that the compressive strength value decreased with an increase in sand/clay ratio.

Dielectric spectroscopy is considered to be a powerful tool because it provides information about the segmental mobility within a polymer. Malström et al.⁷ found three relaxation processes in five generation hyperbranched polyesters with terminal hydroxyl groups⁶ and in three hyperbranched polyesters with the same backbone structure but with different terminal groups (hydroxyl, benzoate, or acetate group⁸), which were the glassy rubber transition (α) and two sub-glass processes (β and γ). The low temperature process (γ) was assigned to motion of the terminal hydroxyl group and the β process to a reorientation of the ester group.

The aim of the present investigation was to systematically study the dielectric and mechanical properties of two polyesters and their mixtures with different ratios. These properties were also studied after the addition of increasing quantities of three different types of fillers (calcium carbonate, clay, and quartz) to the polyesters and their recommended composite.

EXPERIMENTAL

Materials

Reagents

1,2-Propylene glycol, diethylene glycol, 1,6-hexanediol, phthalic anhydride, maleic anhydride, and styrene were obtained from Merck and used without further purification.

Fine grade (0.1 mm) calcium carbonate, Aswan clay, and quartz were obtained from El-Nasr Phosphate Co. with the specifications found in Table I.

Polyesters

Two unsaturated polyester resins based on the reaction of phthalic anhydride (0.5 mol) and maleic anhydride (0.5 mol) with 1,2-propylene glycol (0.525 mol) and 1,6-hexanediol (0.525 mol, PE1) and with diethylene glycol (0.525 mol) and 1,6-hexanediol (0.525 mol, PE2) were prepared by the melt condensation method.⁹

The acid/glycol ratio was 1:1.05 mol. The reaction mixture was heated in an oil bath at 160–200°C until the acid number reached 45 and 56 mg KOH/g for PE1 and PE2, respectively. The polymer resins were purified by dissolving in chloroform, precipitating with light petroleum, and then drying under reduced pressure.

The physical properties of the prepared PE1 and PE2 polyester resins are listed in Table II.

Table II Physical Properties of PE1 and PE2

Property	PE1	PE2
Appearance	Pale yellow	Pale yellow
Specific gravity at 20°C	1.6908	1.5182
Acid value (mg KOH/g)	56	45
Number average molecular weight (\bar{M}_n)	1020	1220
Styrene monomer content in resin (wt %)	30	30

Unsaturated Polyesters with Styrene

The unsaturated polyester resins PE1 and PE2 and a mixture of both of them with different ratios (100/0, 75/25, 50/50, 25/75 and 0/100) were diluted with styrene monomer (70:30 w/w) in the presence of 0.25 wt % methyl ethyl ketone peroxide (MEKP) as an initiator and a 0.25 wt % solution of 8% cobalt naphthenate as a promoter. The polyester/styrene mixtures were cured at room temperature (25°C).

Polymeric Composites

The polymer composites were prepared by mixing different ratios of each of the calcium carbonate, clay, and quartz fillers (20, 40, 60, 70, and 85%) with the polyester/styrene mixture. The prepared composites were left at room temperature until curing occurred.

Methods

The IR spectra were recorded on a Jasco FT/IR 300 E Fourier transform IR (FTIR) spectrometer. The NMR spectrum was run at Jeol Ex-270 NMR spectrometer. The molecular weights of the prepared resins were determined by end-group analysis.¹⁰ Differential scanning calorimetry (DSC) was done with a Perkin-Elmer 7 Series Thermal Analysis System using a heating rate of 5°C/min.

Compressive measurements were carried out according to ASTM D695 using an Instron Universal Testing Instrument (model 1178) on cylindrical rods of 2.5-cm length and 1.3-cm diameter. Tensile measurements were determined using a Universal Testing Machine (fabr. no. 2207/56/15, Veb Leipzig) on cylindrical rods of 10-cm length and 1.3-cm diameter.

Dielectric measurements were carried out in the frequency range of 100 Hz to 100 kHz by using an LCR meter (type AG-411 B, Ando Electric Ltd. Japan). The capacitance (C) and the loss tangent ($\tan \delta$) were directly obtained from the bridge from which the permittivity (ϵ') and dielectric loss (ϵ'') were calculated. The cell used was an 8-mm diameter parallel plate circular condenser made of copper attached to a movable micrometer, which was similar to that designed by Fernandez and Srivastava.¹¹ The cell was calibrated by using standard materials (trolitul, glass, and air) with thicknesses ranging from 1 to 5 mm. For each sample, a relation between the thickness d and its capacitance for the standard materials (C_M) was plotted as a standard curve. The C_M obtained from the standard curves was plotted versus the

known ϵ' for each material for trolitul (2.5), glass (7), and air (1). The relation between the C_M and ϵ' was found to be linear and thus the permittivity corresponding to any measured capacitance could be deduced. Two Teflon samples ($\epsilon' = 2.0$)¹² with different thicknesses were used to check the standard curve. The experimental errors in ϵ' and ϵ'' were 3 and 5%, respectively.

The resistance R was measured directly using a model 6517 Keithley electrometer from which the electrical conductivity (σ) was calculated using the equation

$$\sigma = \frac{d}{RA} \mu\text{S m}^{-1}$$

where d is the thickness of the sample (m) and A is its surface area (m²).

The cell used in these measurements was the one used in the dielectric measurements.

RESULTS AND DISCUSSION

Identification of PE1 and PE2

The structure of the prepared unsaturated polyester resins (PE1 and PE2) was established via IR and NMR spectra. The important bands in their IR spectra from Figure 1 are listed in Table III.

The NMR spectrum of PE1 in Figure 2 indicates the presence of acid residues in the backbone of PE1 by a multiplet between δ 7.8 and 7.4 corresponding to four aromatic protons and a singlet at δ 6.2 corresponding to two olefinic protons.¹³ The figure also indicates the presence of one methine proton of the propylene glycol residue as a broad band between δ 4.8 and 5.4. The group of signals between δ 3.6 and 4.2 are due to the two methylene protons of the propylene residue and four protons of the hexanediol residue. The multiplet between δ 1.2 and 1.8 indicates the presence of three methyl protons of the propylene glycol residue overlapping with eight methylene protons of the hexanediol residue.

Figure 2 also represents the NMR spectrum of PE2. From this figure it is clear that an AB system is present because of the aromatic protons in the phthalic anhydride residue as a multiplet between δ 7.9 and 7.2. The two well-defined doublets at δ 6.8 and 6.2 indicate two olefinic protons. The groups of signals between δ 4.5 and 4.1 are due to six methylene protons of the glycol residue. The multiplet between δ 3.5 and 3.9 is due to six methylene protons of the diethylene glycol resi-

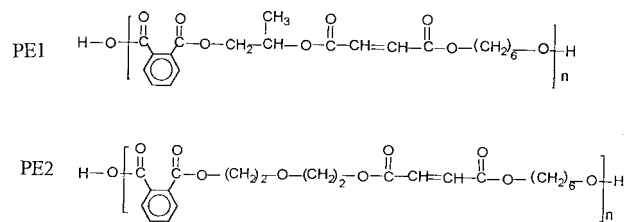
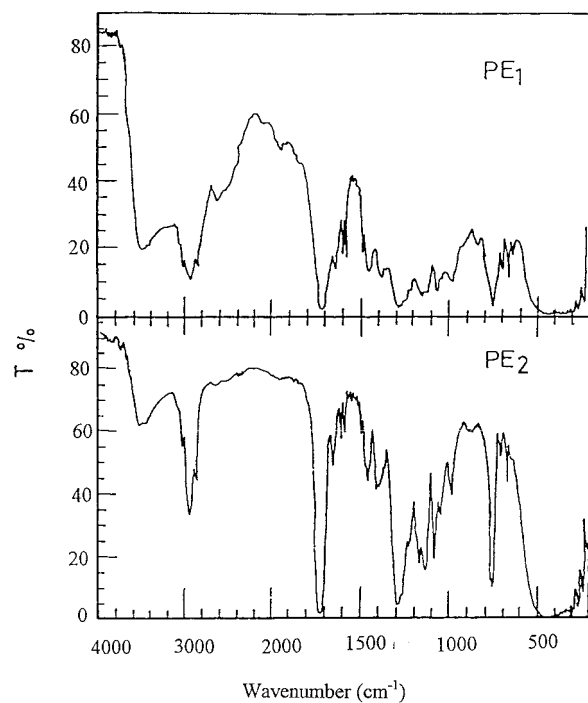


Figure 1 The IR spectrum of polyester resin.

due and the group of signals at δ 1.1–1.7 are due to eight protons of a hexanediol residue.¹³

The thermal behaviors of the styrenated polyester systems (PE1, PE2) and their mixture (50/50 PE1/PE2) as studied by DSC are given in Table IV. From this table it is clear that there was

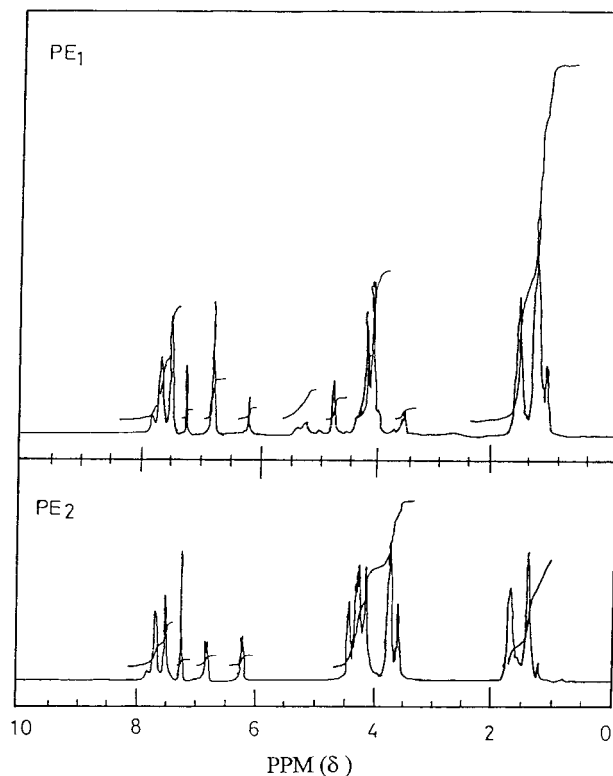


Figure 2 The NMR spectrum of polyester resin.

a broad heat absorption endothermic peak centered on 377 and 387°C for styrenated polyesters PE1 and PE2. This was due to the melting of styrenated polyester. A similar absorption peak was also observed for the 50/50 mixture of PE1/PE2 in the same temperature range.

The ϵ' and ϵ'' for PE1 and PE2 and for different ratios of PE1/PE2 compositions (0/100, 25/75, 50/50, 75/25, and 100/0) were measured in the frequency range of 100 Hz to 100 kHz at room temperature ($\approx 25^\circ\text{C}$). The measured values of ϵ' and ϵ'' are illustrated graphically versus the applied

Table III Main Absorption Bands Detected from IR Spectra

Assignment	Absorption (cm^{-1})	
	PE1	PE2
Stretching OH terminal hydroxyl and stretching OH terminal carboxyl	3580–3440	3580–3440
Stretching CH aromatic and olefinic	2880–2840	2880–2840
Stretching C=O ester	1730 strong	1730 strong, sharp
Stretching C=C olefinic	1640, 1600, 1580	1640, 1600, 1580
Stretching C—O—C— ether	1180	1110
Trans —CH=CH bending	980	980
Cis-olefinic and aromatic residues	660, 700, 780	660, 700, 780

Table IV DSC Results

Styrenated Polyester	T_m (°C)
PE1	377
PE2	387
PE1/PE2(50/50)	383

frequency in Figure 3. From this figure it is clear that the ϵ' decreased with increasing frequency for all investigated samples, which showed anomalous dispersion. On the other hand, the absorption curves relating ϵ'' versus the frequency shown in Figure 3 indicated that more than one relaxation mechanism was present. To the first approximation the analysis of the absorption curves was done in terms of the superposition of two Fröhlich terms, according to the Fröhlich equation¹⁴:

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

where ω is the frequency; P_i is a parameter describing the width of distribution of relaxation times, which equals $\ln(\tau_1/\tau_2)$; $\bar{\tau}$ is the mean relaxation time, which equals $(\tau_1\tau_2)^{1/2}$; ϵ_s is the static permittivity; and ϵ_∞ is the permittivity at infinite frequency. An example of the analyses for PE1, PE2, and 50/50 PE1/PE2 is given in Figure 4. Two relaxation processes were detected. The low frequency was detected at 400 Hz and the corresponding τ and P were 4×10^{-4} s and 2.8, respectively. This region may have been due to either dc conductivity or the Maxwell–Wagner effect or both. This effect should appear at a lower frequency range because of the multiconstituents of the investigated systems. To be certain of this, the electrical conductivity was calculated from the measured resistance and was found to be on the order of $3.0 \times 10^{-14} \mu\text{S m}^{-1}$. The dielectric losses caused by the dc conductivity at the different frequencies (ω) were calculated using the equation¹⁵

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

and were on the order of 5×10^{-4} at low frequency (100 Hz), which was very small when compared with the obtained dielectric losses for these samples at the same frequency (0.04). Thus, it

could be concluded that the absorption region obtained in the low frequency range was attributable to a pure Maxwell–Wagner effect because these polymeric composites were cured with peroxide, which has different permittivity than the polyesters.¹⁶ This region was found to be almost the same for all the samples investigated.

The lower values of σ indicated that the polyesters under investigation were well cured because the conductivity of well-cured polyesters reached a level of $10^{-13} \Omega^{-1} \text{cm}^{-1}$ and the uncured one reached $10^{-9} \Omega^{-1} \text{cm}^{-1}$.¹⁷

In addition to this region, a very distinct higher frequency (70 kHz) process was detected with a relaxation time of 2.3×10^{-6} s and a distribution parameter of one. This region may be attributed to the large aggregates caused by the polarity of the ingredients added to the polyesters during

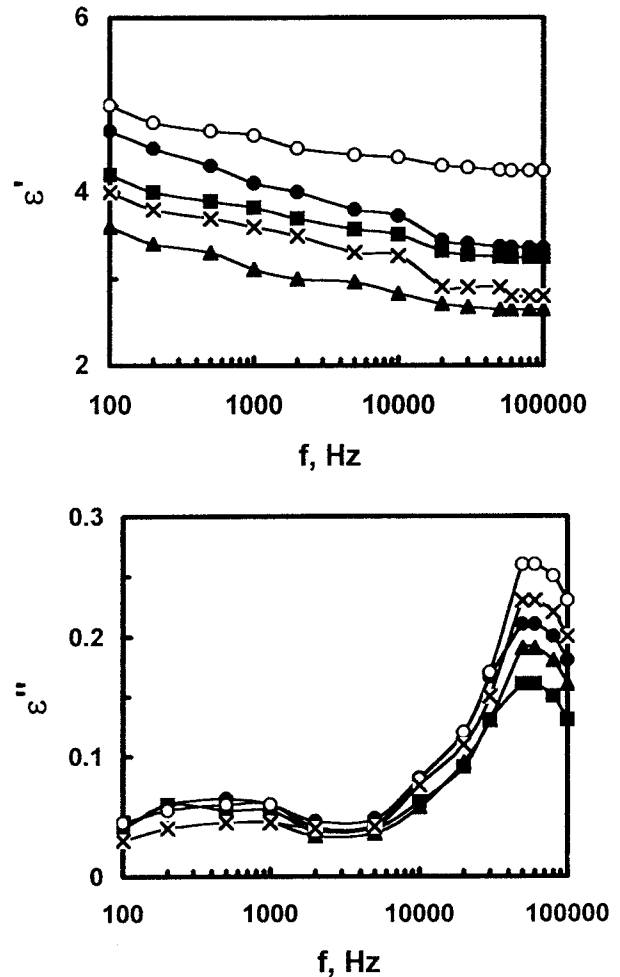


Figure 3 The permittivity (ϵ') and dielectric loss (ϵ'') versus the frequency (f) for styrenated (●) PE1, (▲) PE1/PE2 (75/25), (○) PE1/PE2 (50/50), (■) PE1/PE2 (25/75), and (×) PE2.

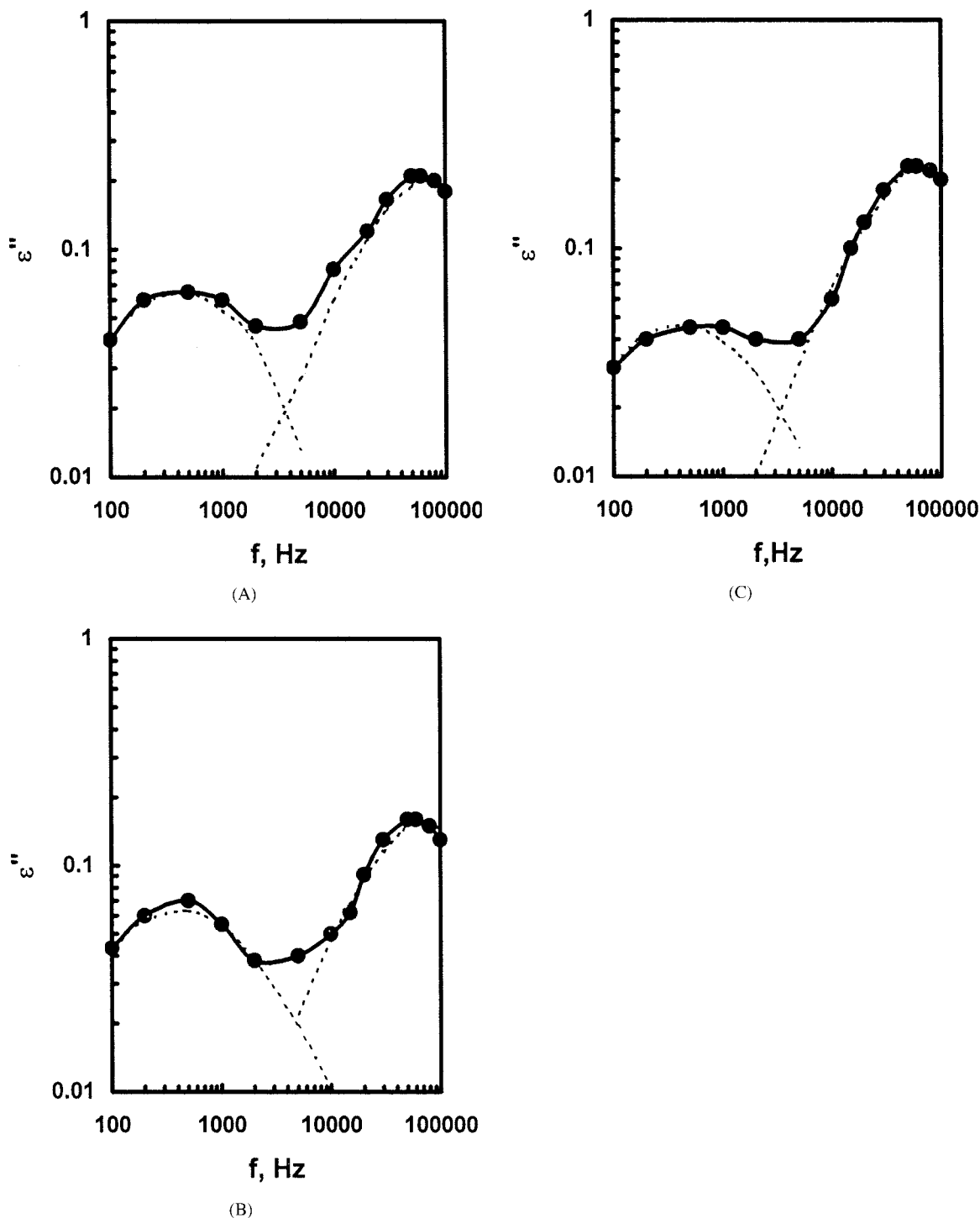
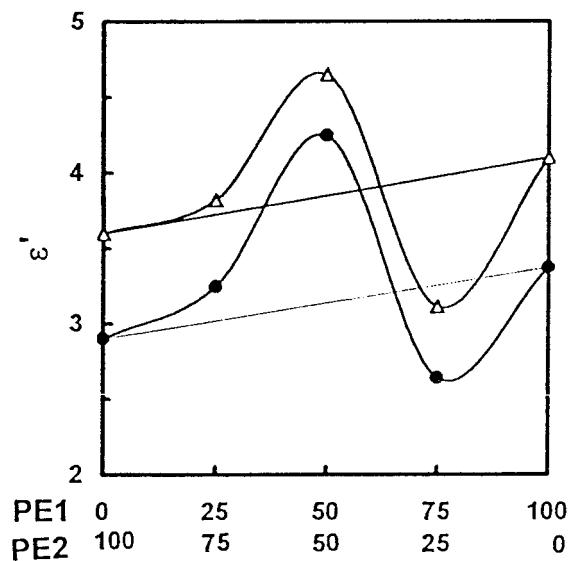


Figure 4 The absorption curves for styrenated (A) PE1, (B) PE1/PE2 (50/50), and (C) PE2, fitting the (—) experimental values (ϵ'') using (---) two Fröhlich terms.

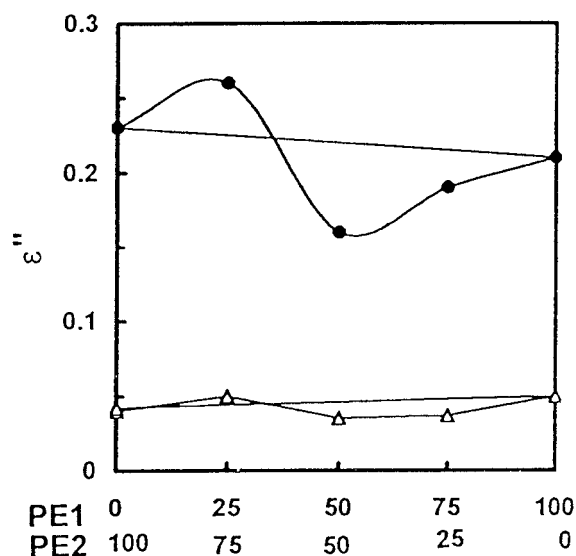
curing such as peroxide.¹⁶ This region was comparable to that found previously.^{18–22} Note that this region was also the same for all the samples that were investigated, which may have been due to the fact that there were no significant changes

in the polarity of the compounds added to either PE1 or PE2.

Figure 5 represents the variation of the ϵ' and ϵ'' at two different frequencies (1 and 50 kHz). From this figure it is clear that the ϵ' at both



PE1/PE2 Content



PE1/PE2 Content

Figure 5 The dependence of the permittivity (ϵ') and dielectric loss (ϵ'') on the styrenated PE1/PE2 content at (Δ) 1 and (\bullet) 50 kHz.

frequencies increased with increasing PE1 content in the composite, reached its highest value at 50% PE1 content (higher than both PE1 and PE2), and then decreased to be less than PE1 itself at 75% PE1. On the other hand it is also clear from the figure that the ϵ'' was much lower at a frequency of 1 kHz than that for 50 kHz for all of the investigated ratios. The values of ϵ'' deviated from the ideal line connecting both indi-

viduals and took an S shape in Figure 5. Thus, the lowest value of ϵ'' given for the sample containing 50% PE1 should be taken into consideration.

From the above investigation we concluded that the system containing 50% PE1 possessed the most promising dielectric properties (highest ϵ' and lowest ϵ''). For this reason this sample and the two individuals (PE1, PE2) were chosen to be loaded with three different types of fillers (calcium carbonate, clay, and quartz), and systematic measurements were carried out.

Polymeric Composites of PE1, PE2, and 50/50 PE1/PE2

The ϵ' and ϵ'' were measured for the two polyesters in addition to the recommended 50:50 composition loaded with different concentrations (0, 20, 40, 60, 70, and 85%) of calcium, clay, and quartz fillers.

The data obtained are graphically illustrated versus the applied frequency in Figures 6–8. From these figures it is clear that the ϵ' decreased with increasing frequency while it increased with increasing filler content to reach its maximum value at 60% for the investigated fillers; then a slight decrease occurred for 70 and 85%. It is also evident that the ϵ' in the whole range of frequencies increased in the order clay > calcium carbonate > quartz, and the 50:50 sample seemed to possess the highest permittivity in all cases.

It was striking to find that the addition of either 60% calcium carbonate or quartz to PE1, PE2, and 50/50 PE1/PE2 led to an increase in the ϵ' values up to 2.5–4 times those of the unfilled samples while it was about 4–4.8 times for the same samples loaded with 60% clay.

The absorption curves relating ϵ'' to the applied frequency in Figures 6–8 indicated that the values of ϵ'' increased with increasing filler content but no pronounced change was noticed when changing the filler type. It was also noted that adding 60% of the fillers increased the ϵ'' by 1.3–2.3 times that of the unfilled samples for the three types of fillers under investigation.

It was of great interest to find that the sample containing 50/50 PE1/PE2 still had the best insulating properties after the addition of fillers because it possessed the highest ϵ' and the lowest ϵ'' for all the fillers and their concentrations.

The absorption curves $\epsilon''(f)$ were analyzed into two Fröhlich terms in the same way as mentioned earlier in the text. An example of the analyses of ϵ'' values for a free 50/50 PE1/PE2 sample in addition to that loaded with 40 and 85% clay is given

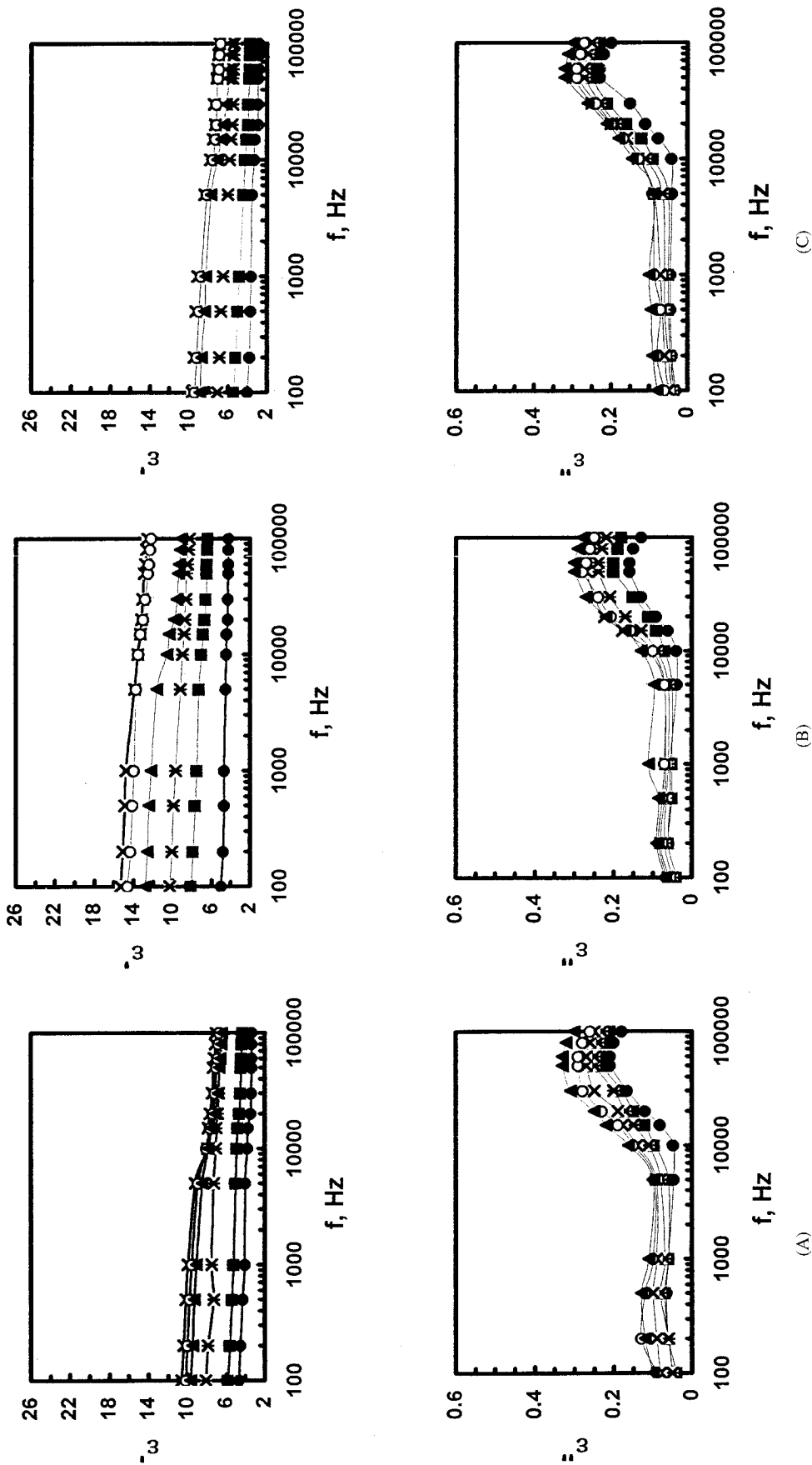


Figure 6 The permittivity (ϵ') and dielectric loss (ϵ'') versus the frequency (f) for styrenated (A) PE1, (B) PE1/PE2 (50/50), and (C) PE2 loaded with (●) 0, (■) 20, (×) 40, (○) 60, (◻) 70 and (▲) 85% calcium carbonate.

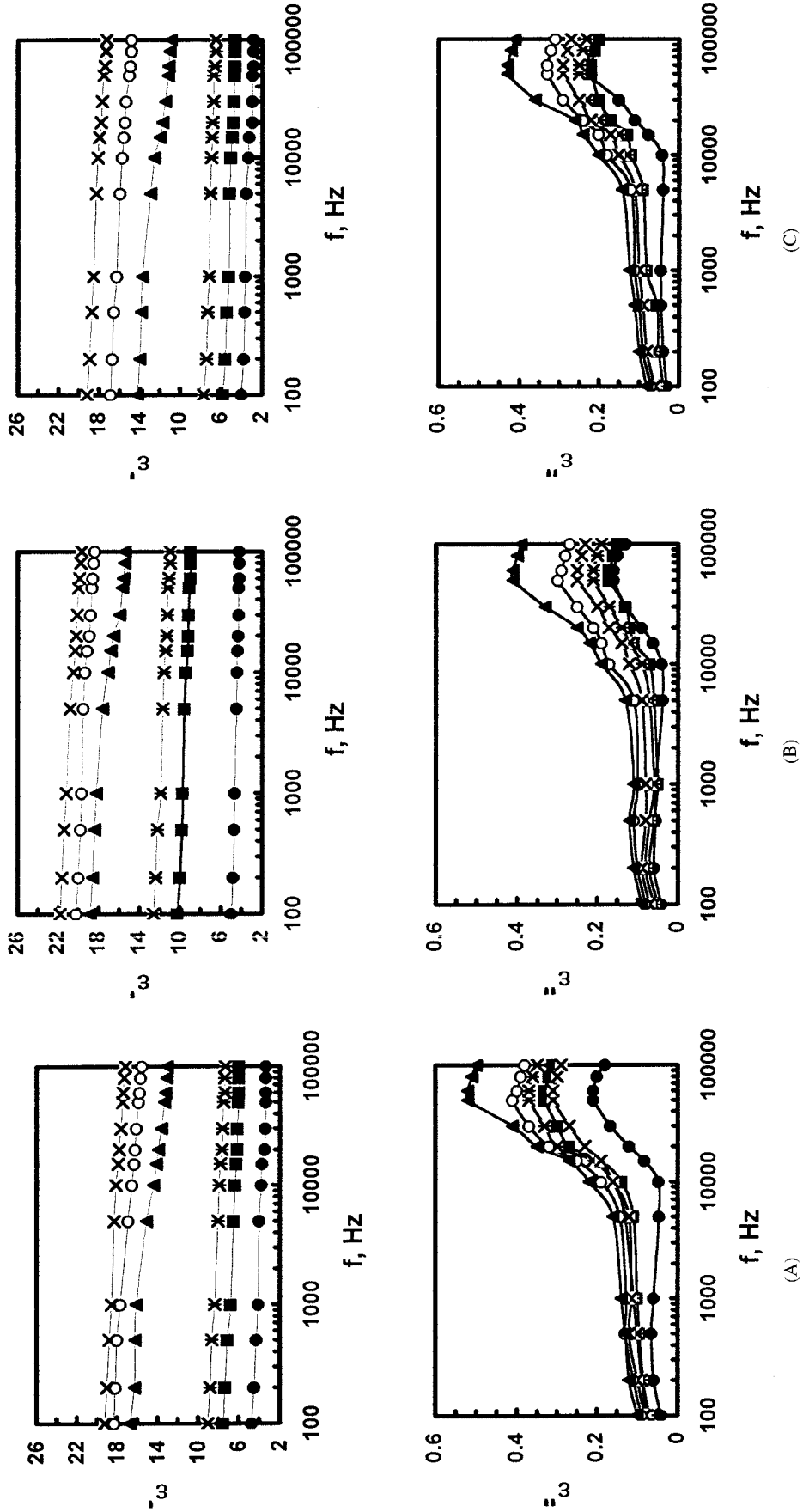


Figure 7 The permittivity (ϵ') and dielectric loss (ϵ'') versus the frequency (f) for styrenated (A) PE1, (B) PE1/PE2 (50/50), and (C) PE2 loaded with (●) 0, (■) 20, (*) 40, (×) 60, (○) 70, and (▲) 85% clay.

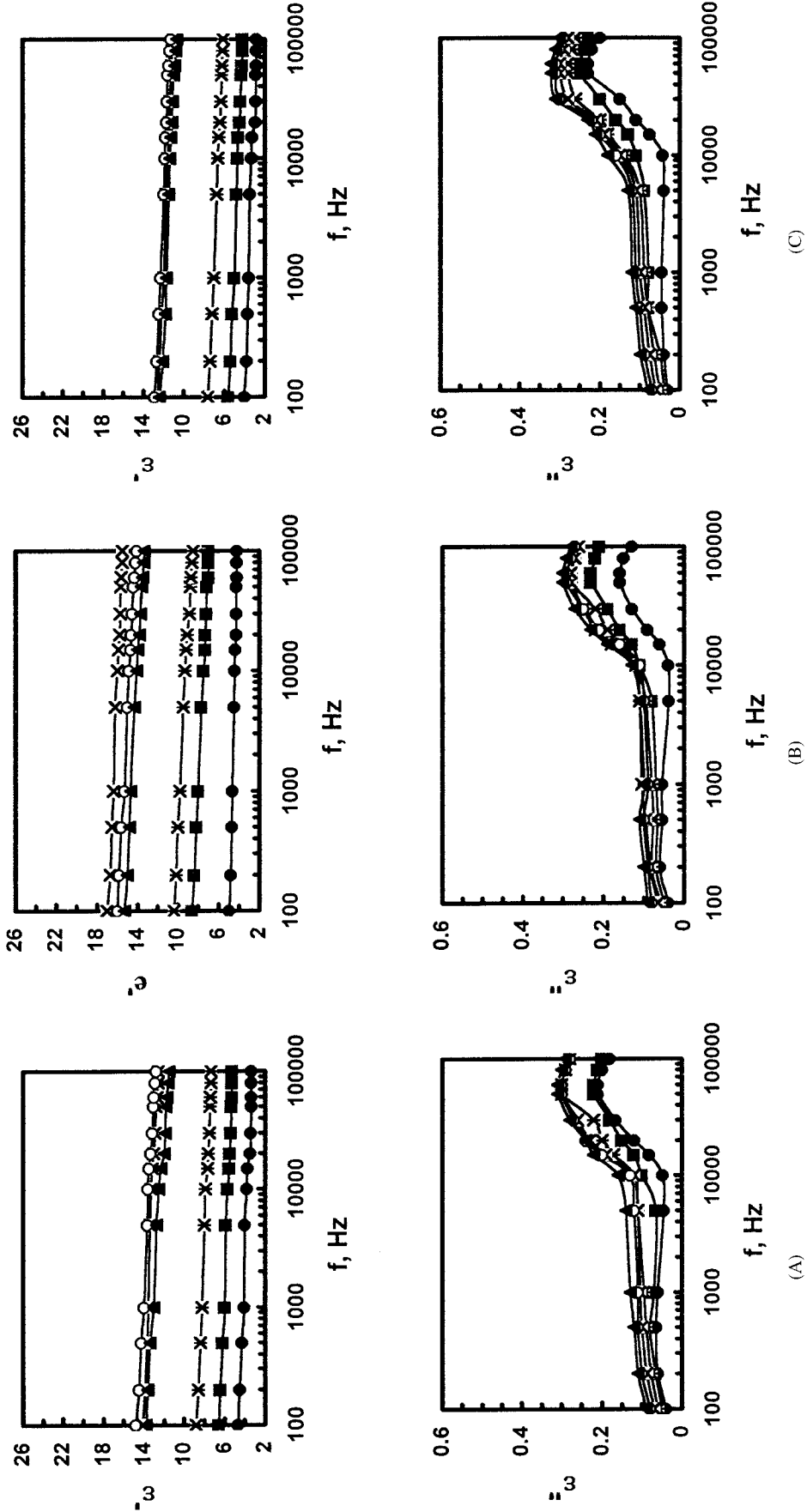


Figure 8 The permittivity (ϵ') and dielectric loss (ϵ'') versus the frequency (f) for styrenated (A) PE1, (B) PE1/PE2 (50/50), and (C) PE2 loaded with (●) 0, (■) 20, (*) 40, (×) 60, (○) 70, and (▲) 85% quartz.

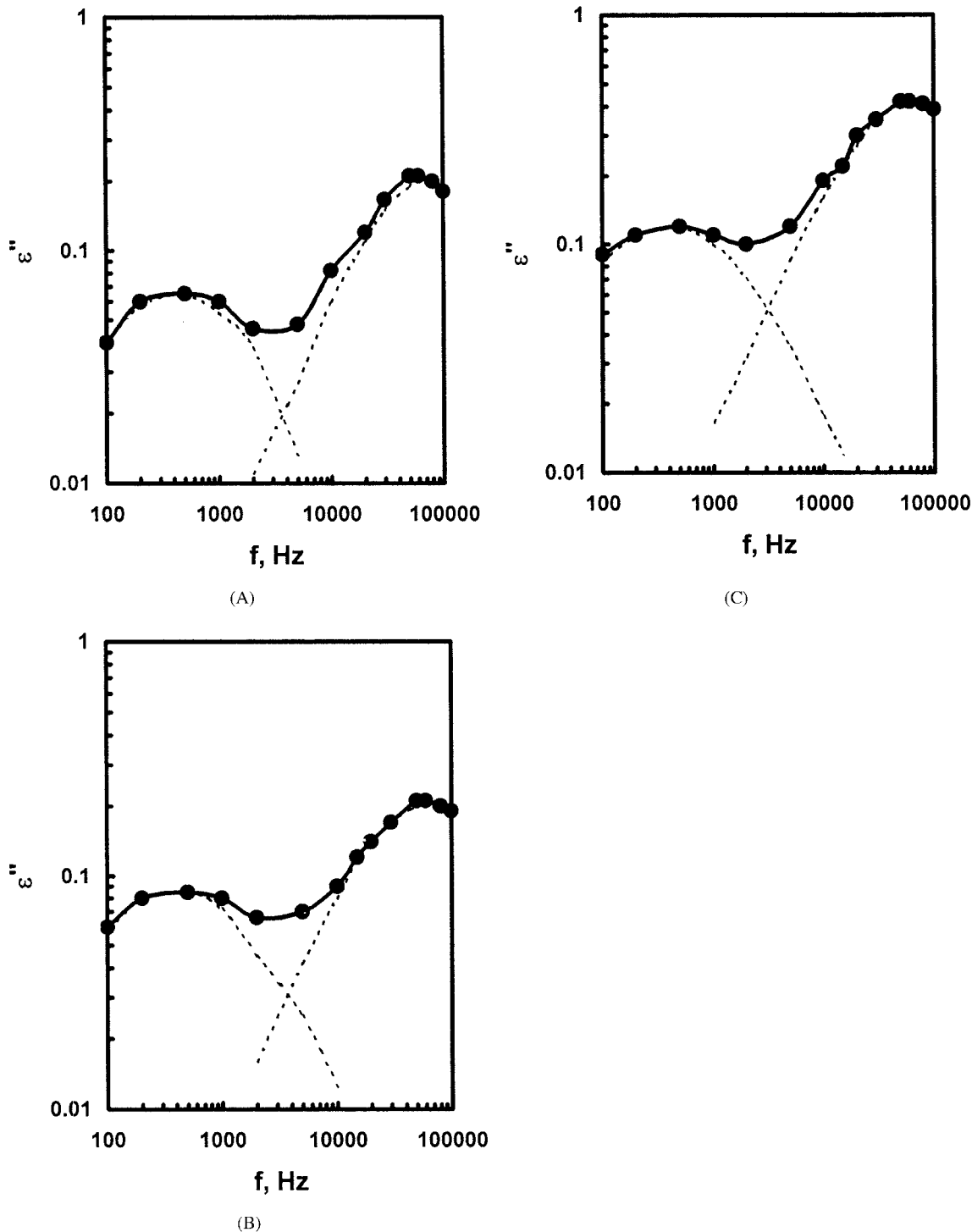


Figure 9 The absorption curves for styrenated PE1/PE2 (50/50) with (A) 0, (B) 40, and (C) 85% clay, fitting the (—) experimental values (ϵ'') using (---) two Fröhlich terms.

in Figure 9. The lower frequency absorption region may have been due to the Maxwell-Wagner effect, which usually appears in such a range of frequencies because the values of the σ were very small (on the order of $10^{-14} \mu\text{S m}^{-1}$); conse-

quently, the losses due to the dc conductivity were negligible. The value of the relaxation time associated with that region was similar to that detected in the free samples ($4 \times 10^{-4} \text{ s}$). Moreover, this region was comparable to that found previ-

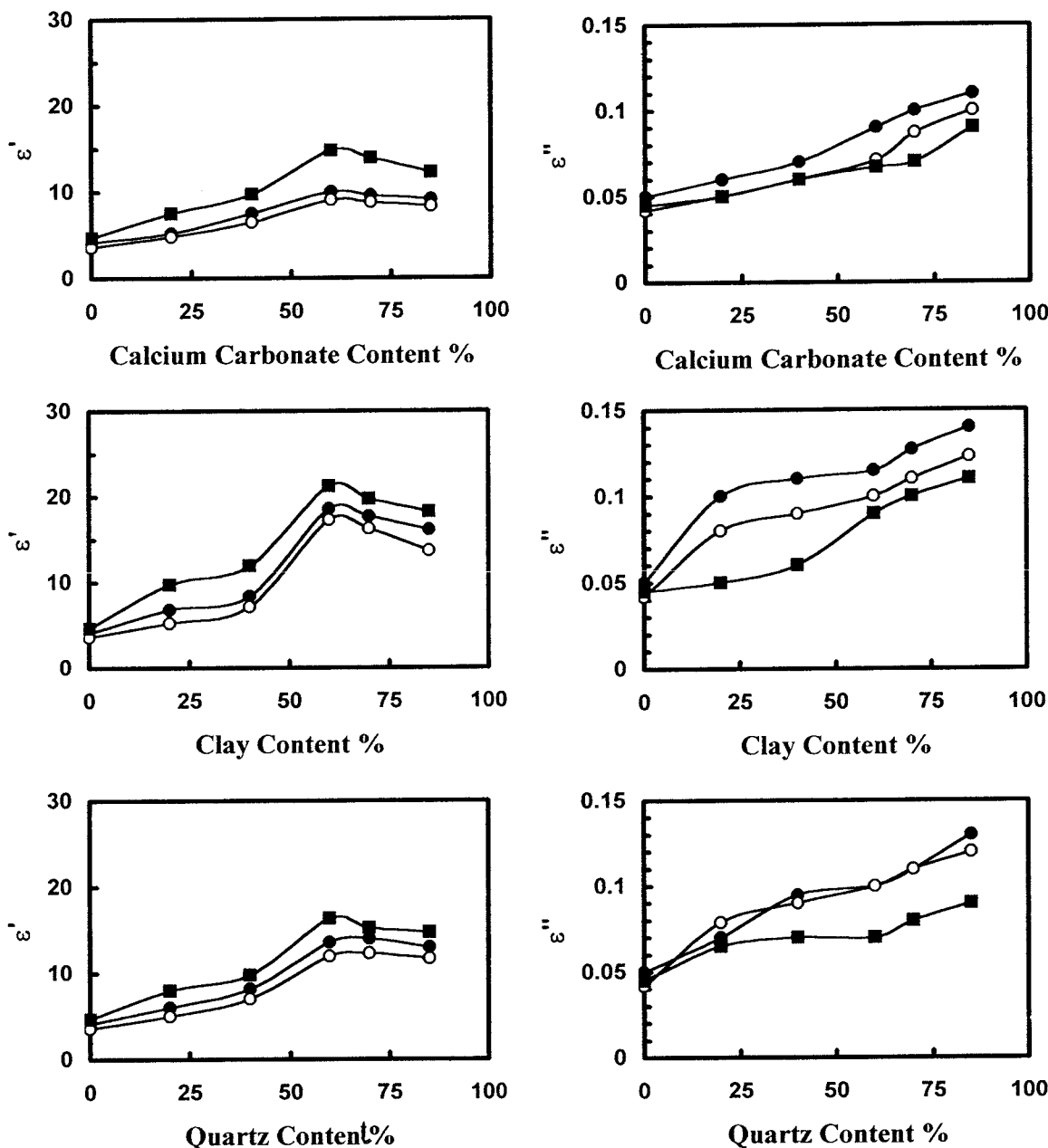


Figure 10 The variation of the permittivity (ϵ') and dielectric loss (ϵ'') at 1 kHz versus the filler content for styrenated (●) PE1, (○) PE2, and (■) PE1/PE2 (50/50).

ously in an ethylene-propylene-diene monomer/chloroprene rubber blend loaded with white and black filler.²⁰

In addition to this region, another distinct relaxation process was detected at about 70 kHz. This region could be attributed to the large aggregates caused by the polarity of the ingredients added to the polyesters during curing such as peroxide.¹⁶ Figure 9 shows that the position of the maximum frequencies was not changed by the addition of the filler, whatever its concentration.

This suggested that there was no chemical interaction between these fillers and the investigated polyesters.^{21,22}

The data in Figure 10 represent the variation of ϵ' and ϵ'' versus the percentage of filler (calcium carbonate, clay, and quartz) at a frequency of 1 kHz. From this figure the following facts were noted:

- The values of ϵ' and ϵ'' were increased by increasing the filler content and reach their

Table V Compressive Strength and Tensile Strength Results

Styrenated Polyester	Filler	Calcium Carbonate		Clay		Quartz	
		0	60%	70%	60%	70%	60%
Compressive Strength (kg/cm ²)							
PE1	1078	703	917	848	—	1074	942
PE2	1168	490	414	728	704	515	613
PE1/PE2 (50/50)	1055	606	477	942	832	885	747
Tensile Strength (kg/cm ²)							
PE1	83	188	75	113	—	128	42
PE2	151	64	40	66	30	68	98
PE1/PE2 (50/50)	181	106	30	30	16	124	79

maximum value at 60%, and then a slight decrease in ϵ' occurred at 70 and 85%.

- The values of ϵ' increased in the order 50/50 PE1/PE2 > PE1 > PE2 while ϵ'' increased in the order PE1 > PE2 > 50/50 PE1/PE2.
- All the polyesters that were loaded with clay with different concentrations showed the highest values of ϵ' .
- The sample containing 50/50 PE1/PE2 possessed the highest ϵ' and the lowest ϵ'' for the three investigated fillers with different concentrations.
- Styrenated polyesters loaded with clay, which is a local filler, possessed the highest ϵ' and slightly higher ϵ'' compared to those for calcium carbonate and quartz.
- From the above investigation it could be concluded that the sample containing 50/50 PE1/PE2 loaded with 60–70% clay possessed the most promising dielectric properties because it had the highest values of ϵ' .
- Thus, clay, which is a cheap and available filler, could be used with the investigated polyesters in a high concentration (60–70%) to obtain an economical end product, which possessed promising dielectric properties.

A comparative study was done on the three types of fillers (calcium carbonate, clay, and quartz) with the two recommended concentrations (60 and 70%). The compressive strength and the tensile strength values were measured for such samples and listed in Table V.

The highest compressive strength values were obtained for PE2 when compared with those de-

tected for PE1 and 50/50 PE1/PE2. The results depicted in Table V clearly indicate that, among the three fillers used, quartz and clay imparted the highest compressive values to the polymer composites when the filler content was 60% in PE1 and 50/50 PE1/PE2.

The results listed in Table V reveal that the highest value for the tensile strength was obtained with 50/50 PE1/PE2 followed by PE2, and the lowest one was obtained for PE1. After the addition of the different types of fillers, calcium carbonate and quartz composites (60% filler) in PE1 and 50/50 PE1/PE2 imparted the highest tensile strength values as compared with clay.

The compressive and tensile strength values for the polymeric composite containing 70% clay in PE1 could not be tested because noncompact samples were obtained.

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