

Synthesis and Characterization of Unsaturated Polyester/Carborundum Composites

Jeannette N. Asaad

Polymers and Pigments Department, National Research Centre, Cairo, Egypt

Correspondence to: J. N. Asaad (E-mail: na.jeannette@hotmail.com)

ABSTRACT: Two unsaturated polyesters, one based on phthalic anhydride PE_P and the other based on isophthalic acid PE_I, were synthesized. The chemical structure of the two polyesters was characterized by IR and ¹H and ¹³C NMR spectroscopy. The effect of styrene concentration on the curing of polyesters was also studied. It has been found that the percent of polyester/styrene (70/30 wt %) gave the highest percent of curing. Different concentrations of carborundum (0–70 wt %) were used to prepare polyester composites. A comparative study was done on the properties of the two prepared polyesters PE_P and PE_I and their composites in term of their thermal, mechanical, electrical, and physical properties. The results indicate that the polyester based on isophthalic acid (PE_I) and its composites gave higher compressive strength values and lower water absorption than those based on phthalic anhydride (PE_P). The presence of carborundum improved the thermal stability than the cured polyesters and electrical properties. © 2012 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* 129: 1812–1819, 2013

KEYWORDS: polyesters; composites; dielectric properties; properties and characterization; thermal properties

Received 10 August 2012; accepted 23 November 2012; published online 24 December 2012

DOI: 10.1002/app.38875

INTRODUCTION

Unsaturated polyesters are become increasingly popular and widely produced industrially because of their low cost, easy processing, and extremely versatile in properties and applications.^{1–3} They are usually prepared by polycondensation, typically of a dihydroxyl compound or a mixtures of dihydroxy compounds (diols) with maleic anhydride (MA) and/or together with other dicarboxylic acids, such as aromatic or aliphatic dicarboxylic acids under elevated temperature.⁴ The most traditional composition is MA, *o*-phthalic anhydride, and 1,2-propanediol, which are cheap raw materials. Other common raw materials are fumaric acid, isophthalic anhydride, terephthalic acid, adipic acid, ethylene glycol, diethylene glycol, dipropylene glycol, neopentyl glycol, and bisphenol A. The properties of the final product can be varied almost endlessly by changing the composition of the unsaturated polyester using these raw materials.^{5–8} Generally, aromatic groups improve the hardness and the stiffness, whereas aliphatic chain components increase the flexibility. The incorporation of isophthalic acid creates a high-molecular-weight resin with good chemical, water, and thermal resistance and good mechanical properties.⁸

The unsaturated polyester prepolymer is finally blended with styrene to form a reactive resin solution, in which the resin is sent to the end-user. The styrene acts both as a crosslinking

agent and as a viscosity reducer so that the resin can be processed in conventional unsaturated polyesters. Moreover, these resins can be formulated and chemically tailored to provide properties and process compatibility that meet the requirements of specific end user applications.

Unsaturated polyester resins are one of the most important thermoset materials used in composites industry. They have made significant structural and performance advances,^{9,10} which resulted in their broader application in automotive, building, construction, electrical, and marine industries.^{11,12} Despite those many advantages, such as ease of fabrication, low production costs, easily controllable, and fast cure processes at room temperature, transparency,¹³ the typical polyesters have some disadvantages. They are not tough enough, highly flammable, and their impact strength is lower than of other thermoset polymers like epoxy.¹⁴

Therefore, their properties are often improved and modified in two commonly used methods: physical blending and compounding and chemical modification.¹⁵

The improved performance of polyesters and their composites in industrial and structural applications by the addition of filler materials has shown great promise and so has lately been a subject of considerable interest. The inclusion of inorganic fillers into polymers for commercial applications is primarily aimed at

Table I. Physical Properties of Phthalic (PE_p) and Isophthalic Polyester (PE_i)

Property	PE _p	PE _i
Appearance	Yellow	Yellow
Acid value, mg KOH/g	35	30
Specific gravity at 20°C	1.25	1.11
M. wt calculated	1603	1870

the cost reduction and stiffness improvement.^{16–19} These engineering composites are desired due to their low density, high corrosion resistance, ease of fabrication, and low cost.^{20,21}

Carborundum, known as silicon carbide (SiC), is filler that was originally produced by a high temperature electro-chemical reaction of sand and carbon with chemical formula SiC. It is one such ceramic material that has the potential to be used as filler in various polymer matrices. It is used in abrasives, refractory, ceramics, and numerous high performance applications. The material can also be made an electrical conductor and has applications in resistance heating, flame igniters, and electronic components. Structural and wear applications are constantly developing.²²

In this work, a comparative study was done on the properties of two prepared polyesters PE_p and PE_i based on phthalic anhydride and isophthalic acid, respectively. Their composites filled with different concentrations of Carborundum were investigated with respect to thermal, mechanical, electrical, and physical properties,

EXPERIMENTAL

Materials

The pure grades of dipropylene glycol, phthalic anhydride, isophthalic acid, and MA were obtained from Merck Co (Germany). Also, styrene monomer was reagent grade from Merck, Darmstadt, Germany and used as received without further purification. Fine chemicals carborundum powder 400 mesh was obtained from Market Harborough, Leicestershire, and L.E 16 9EJ. U.K

Synthesis of Polyesters

Two unsaturated polyesters based on the reaction of phthalic anhydride (0.5 mol) (PE_p), or isophthalic acid (0.5 mol) (PE_i) each with MA (0.5 mol) and dipropylene glycol (1.05 mol) were prepared by melt condensation. The acid/glycol ratio was 1 : 1.05 mol. The reaction mixture was stirred under a continuous nitrogen flow for 6 h and the temperature was increased from 150 to 220°C in an oil bath. The acid value was monitored during the reaction and determined by end-group analysis of samples collected every hour. The acid number reached 35 and 30 mg KOH/g for PE_p and PE_i, respectively, at the end of the reaction. The prepared polyesters resins were heated under vacuum for 30 min to remove water and the unreacted monomers. The physical properties of the prepared polyester resins are listed in Table I.

Curing of the Prepared Unsaturated Polyesters

The resins were cross-linked with different percentage of styrene monomer (30, 40, and 50 wt %) in the presence of methyl ethyl ketone peroxide as initiator (1 wt %) and cobalt naphthenate as accelerator (0.25 wt %). Curing occurred after the mixtures were left at room temperature overnight followed by 2 h at 80°C in a thermostated oven until complete curing occurred.

Percent of Curing

Calculated weight of styrenated polyesters was soaked in chloroform for 7 days after that the specimen was dried and weighed. The percent of curing was calculated according to the following equation:

$$\% \text{ of curing} = \frac{\text{weight after soaked in chloroform}}{\text{weight before soaking}} \times 100$$

Polyester Composites

Polyester composites were prepared by mixing different ratios (30, 40, 50, 60, and 70 wt %) of carborundum powder (400 mesh) with both polyester/styrene mixture (70/30 wt %). The prepared samples of appropriate shapes for various measurements were molded and cured at the same conditions.

Techniques

IR spectra were recorded on a Jasco FT/IR 300 E Fourier transform infrared spectrometer (Tokyo, Japan).

The nuclear magnetic resonance ¹H and ¹³C NMR spectrum was run at 260 MCPS on a Jeol Ex-270 NMR spectrometer (Tokyo, Japan).

Compression measurements were carried out according to ASTM D 695- 05 using Instron Universal Testing Instrument Model 1178.

The physical properties were tested and calculated according to ANSI/ASTM C20-00 (2010). The volume of open pores and the volume of impervious portions of the specimens were calculated as follows: Volume of open pores, cm³ = *W* – *D*.

Volume of impervious portion, cm³ = *D* – *S*. where *W* = saturated weight; *D* = dry weight; *S* = suspended weight.

The apparent porosity (*P*) expresses the percent relationship of the volume of the open pores of the specimen to its exterior volume and was calculated as follows:

$$P\% = \frac{W - D}{V} \times 100$$

where *W* = saturated weight; *D* = dry weight; *V* = exterior volume.

The water absorption (*A*), expresses a percent relationship of water absorbed to the weight of the dry specimen and was calculated as follows:

$$A\% = \frac{W - D}{D} \times 100$$

where *W* = saturated weight; *D* = dry weight.

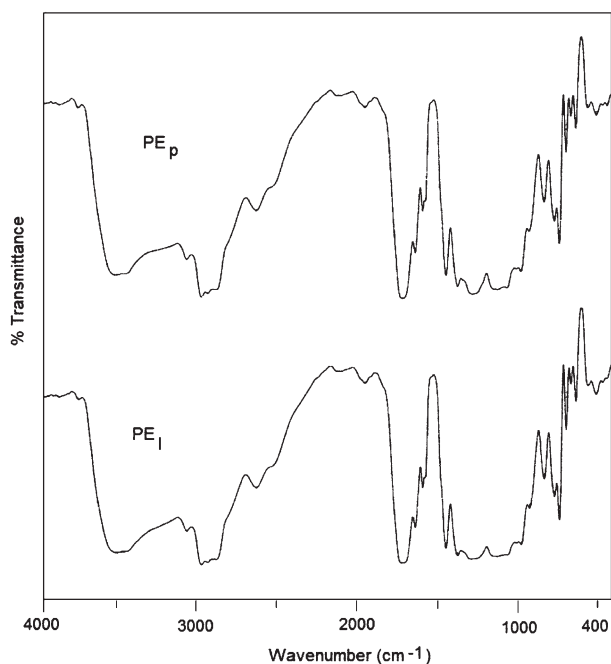


Figure 1. IR spectrum of the prepared polyester resins (PE_p and PE_I).

The apparent specific gravity (T) is a measure of that portion of the test specimen which is impervious to boiling water and was calculated as follows:

$$T = \frac{D}{D - s}$$

where D = dry weight; S = suspended weight.

The bulk density (B) in grams per cubic centimeter of a specimen is the quotient of its dry weight divided by the exterior volume, including pores and was calculated as follows:

$$B = \frac{D}{V} \text{ g / cm}^3$$

where D = dry weight; V = exterior volume.

Thermogravimetric analysis (TGA) was performed using Perkin-Elmer, TGA 7 (USA) instrument. The rate of heating was 10°C/min up to 700°C under nitrogen atmosphere.

Dielectric measurements were performed in the frequency range of 100 Hz–100 kHz by using an LCR meter type AG-411 B (Ando electric Ltd., Japan). The measurements were carried out at room temperature $\sim 25^\circ\text{C}$. The capacitance C , the loss tangent $\tan \delta$, and the resistance R were obtained directly from the bridge from which the permittivity ϵ' , dielectric loss ϵ'' , and conductivity σ were calculated.

A guard ring capacitor type NFM/5T Wiss Tech. Werkstaten (WTW) GMBH Germany was used as a measuring cell. The cell was calibrated using standard materials²³ and the experimental error in ϵ' and ϵ'' was found to be $\pm 3\%$ and $\pm 5\%$, respectively.

RESULTS AND DISCUSSION

Characterization of the Prepared Polyesters

The molecular structures of the prepared polyesters PE_p and PE_I were identified using IR, ^1H and ^{13}C NMR spectroscopy. The

Table II. The Main Peak Assignments Detected from IR Spectra

Assignments	Peak (cm ⁻¹)	
	PE _p	PE _I
O[–]H unassociated, associated with alcohol or carboxyl	3507	3504
Aromatic and aliphatic C[–]H stretching	2977	2977
C[=]O ester stretching	1725	1732
[–]C[=]C[–] olefinic and aromatic stretching	1646–1600	1646–1600
[–]C[–]O[–]C[–] ether Stretching	1132	1136
Trans [–]CH[=]CH[–] bending	984	985
Cis-olefinic and aromatic residue	778–747	778–747

IR spectra of PE_p and PE_I (Figure 1) were found to be quite similar and illustrate the presence of important linkage such as ester groups, olefinic double bond and other characteristic peaks. Peak assignments in the IR spectra of the prepared resins are shown in Table II.

The ^1H and ^{13}C NMR spectra of PE_p and PE_I exhibit well resolved signals and indicate that the two resins have similar features.

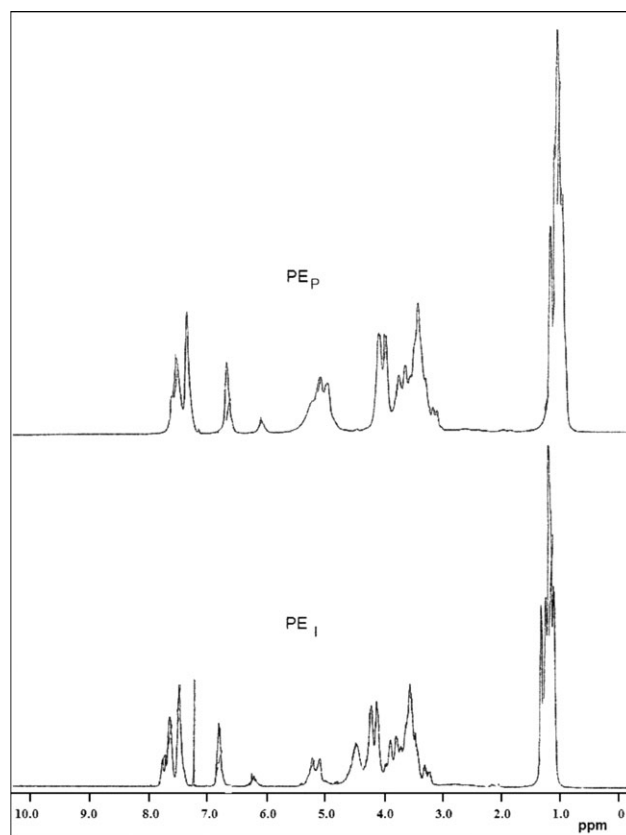


Figure 2. ^1H NMR spectrum of the prepared polyester resins (PE_p and PE_I).

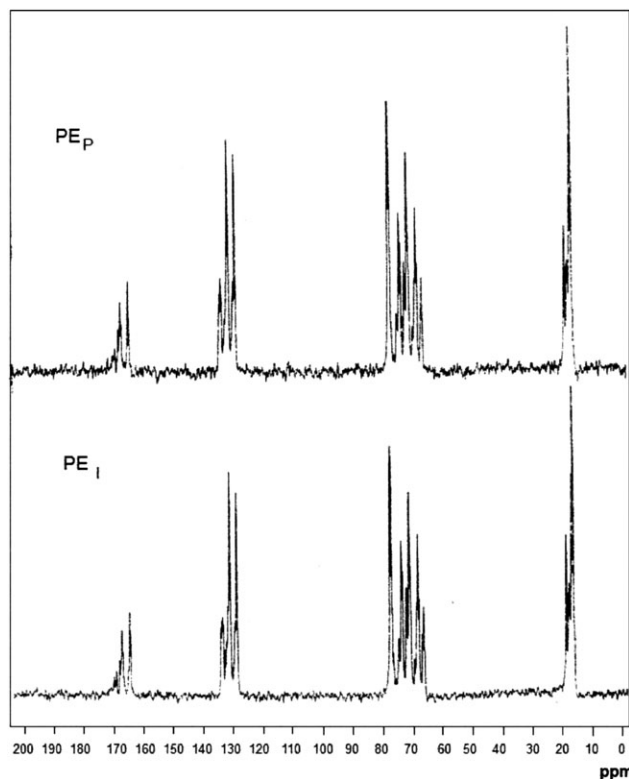


Figure 3. ^{13}C NMR spectrum of the prepared polyester resins (PE_p and PE_i).

From the ^1H NMR spectra shown in Figure 2, it is possible to distinguish the methyl, methylene and methine protons of dipropylene glycol at δ 1–1.5, 3.2–4.8, and 5.3 ppm, respectively. The asymmetrical signals at δ 6.6–7 ppm seem to be two ill-defined doublets for the olefinic protons in the maleic-fumaric acid residue. The signals between δ 7.4 and 8 ppm are characteristic of the aromatic protons of phthalic anhydride and isophthalic acid in the polyester backbone. The signal appears at δ 7.3 ppm which is due to the proton of solvent (chloroform).

In ^{13}C NMR spectra (Figure 3), the methyl group of propylene glycol appears at 20 ppm. The signals assigned to aliphatic carbons of the glycol moiety are located in the region between 65 and 80 ppm. The groups of the signals between 128 and 138 ppm are most probably due to the aromatic carbons of the acid moieties. The carbonyl carbon can be assigned to the region between 165 and 170 ppm.

Percent of Curing

The percent of curing of the two prepared polyesters containing different styrene contents was calculated and presented in Table III.

Table III. Percent of Curing of Unsaturated Polyesters

Styrene wt %	Percent of curing	
	PE_p	PE_i
30	94.47	98.50
40	93.07	93.70
50	90.09	90.60

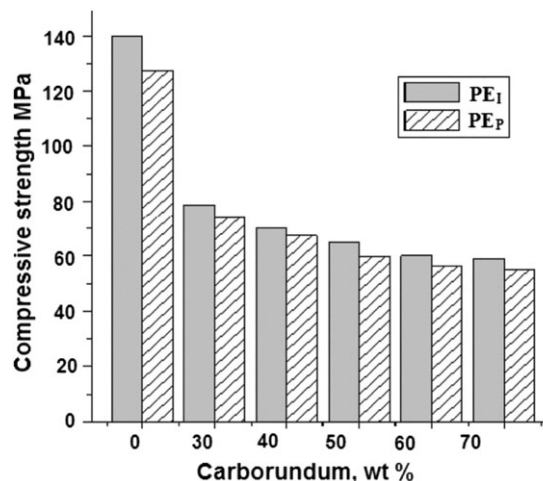


Figure 4. Compressive strength values of two styrenated polyester (PE_p and PE_i) and their composites.

It is clear that, the curing percent decreases with increasing styrene content. A possible explanation of this behavior is the diffusion limitation. The increase of styrene content can cause additional production of free radicals which leads to the decrease of time to reach the gel point. When the system reaches the gel-point (gelation), it is considered as a high molecular weight network and consequently, the viscosity reaches infinity and the final conversion occurs as a diffusion-controlled process.²⁴

It was noticed that for both resins the samples containing 30 wt % of styrene exhibit the highest percent of curing. Therefore, this percent was used to prepare all cured samples with appropriate molds for different tests. Also, the polyester based on isophthalic acid (PE_i) has higher percent of curing than the polyester based on phthalic anhydride (PE_p) at this percent.

Mechanical Properties

The mechanical properties of PE_p and PE_i as a function of filler content were evaluated through compressive strength test and the results are shown in Figure 4. It was found that, the compressive strength values of the cured polyester resins were comparatively higher than that of the filled polyester composites. Generally, a lowering in the compressive strength values was observed as the filler content increased.^{17,25} This may be due to the fact that the incorporation of the filler reduces the volume fraction of the matrix and subsequently lowered the ability of the sample to absorb energy or distribute stress efficiently. It has been reported that the mechanical properties are dependent on the relative volumes of any given polymer matrix material and fillers.^{4,26,27} Also, there are another two reasons that may explain why the compressive strength of these filled composites was lower than those of the unfilled ones. One is that the chemical reaction at the interface between carborundum particles and the matrix may be too weak to transfer the compressive stress; the other is that the corner points of the irregular-shaped carborundum particles result in stress concentration in the polyester matrix.²² It is interesting to note that among the two polyesters, the samples based on isophthalic acid (PE_i) impart

Table IV. Physical Properties of PE_p Composites

Properties	Carborundum wt %					
	0	30	40	50	60	70
Exterior volume V (cm ³)	3.71	6.22	5.54	5.54	4.97	5.27
Volume of impervious portion (cm)	3.70	6.20	5.54	5.53	4.96	5.2
Volume of open pores (cm ³)	0.009	0.01	0.01	0.007	0.005	0.004
Apparent porosity P (%)	0.23	0.20	0.17	0.128	0.098	0.08
Water absorption A (%)	0.19	0.17	0.14	0.11	0.08	0.07
Apparent specific gravity T	1.20	1.15	1.16	1.169	1.2	1.17
Bulk density B (g/cm ³)	1.20	1.147	1.16	1.167	1.19	1.17

Table V. Physical Properties of PE_i Composites

Properties	Carborundum wt %					
	0	30	40	50	60	70
Exterior volume V (cm ³)	5.07	6.003	7.34	4.46	4.50	5.563
Volume of impervious portion (cm)	5.06	5.99	7.33	4.47	4.50	5.56
Volume of open pores (cm ³)	0.008	0.007	0.008	0.004	0.003	0.003
Apparent porosity P (%)	0.16	0.12	0.106	0.10	0.069	0.054
Water absorption A (%)	0.13	0.101	0.094	0.085	0.056	0.046
Apparent specific gravity T	1.17	1.15	1.13	1.19	1.22	1.17
Bulk density B (g/cm ³)	1.16	1.15	1.13	1.19	1.22	1.17

higher compressive strength values than those based on phthalic anhydride (PE_p).

Physical Characteristics of Polyester Composites

The physical properties of the polyester matrices are important because they strongly influence the performance of the composites. Table I lists a comparative physical property of the two synthesized resins that used in the preparation of the polyester composites.

Some physical properties of the prepared polyester/carborundum composites were evaluated and the results have been repre-

sented in Tables IV and V. It is clear from the data obtained that acceptable values for exterior volume, volume of impervious portion, volume of open pores, apparent porosity, water absorption, apparent specific gravity, and bulk density were reached.

These composites were found to have small volume of open pores, low water absorption, and low apparent porosity. The values of these properties were slightly decreased with increasing the filler content in the polymeric composites. The low water

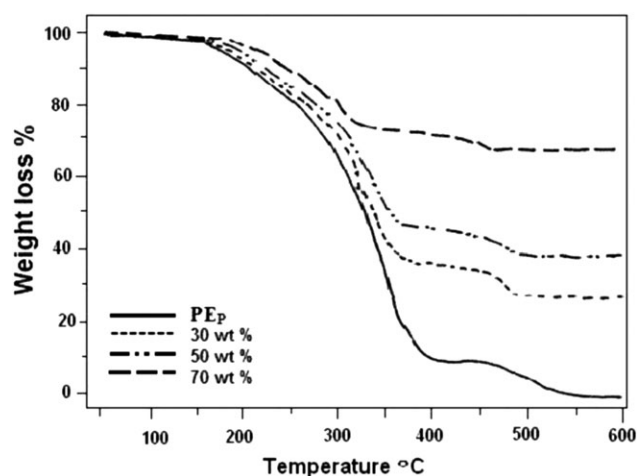
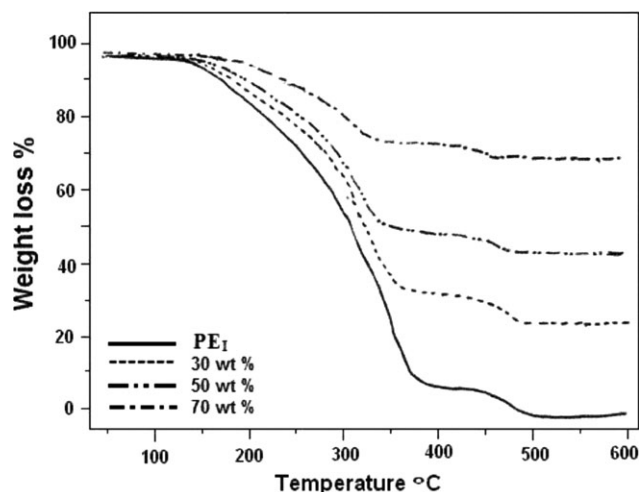
**Figure 5.** TGA curves of styrenated polyester (PE_p) and its composite filled with 30, 50, and 70 wt% of carborundum.**Figure 6.** TGA curves of styrenated polyester (PE_i) and its composite filled with 30, 50, and 70 wt% of carborundum.

Table VI. TGA Data of PE_p/Composites

Carborundum wt %	T ₀ (°C)	T ₅₀ (°C)	Char yield % at 600°C
0	150	325	2
30	155	340	30
50	160	360	42
70	180	-	70

Table VII. TGA Data of PE_i/Composites

Carborundum wt %	T ₀ (°C)	T ₅₀ (°C)	Char yield % at 600°C
0	150	325	2
30	160	340	30
50	175	490	48
70	200	-	75

absorption of these investigated composites would suggest that these polyester composites can be used in many industrial applications, since they can prevent the intrusion of water and salts that could cause corrosion of the installation.²⁸

Thermogravimetric Analysis

The thermograms of the two styrenated polyesters PE_p and PE_i and their composites filled with 30, 50, and 70 wt% of carborundum are shown in Figures 5 and 6. Two degradation steps were observed at 150 and 350°C for all samples, corresponding

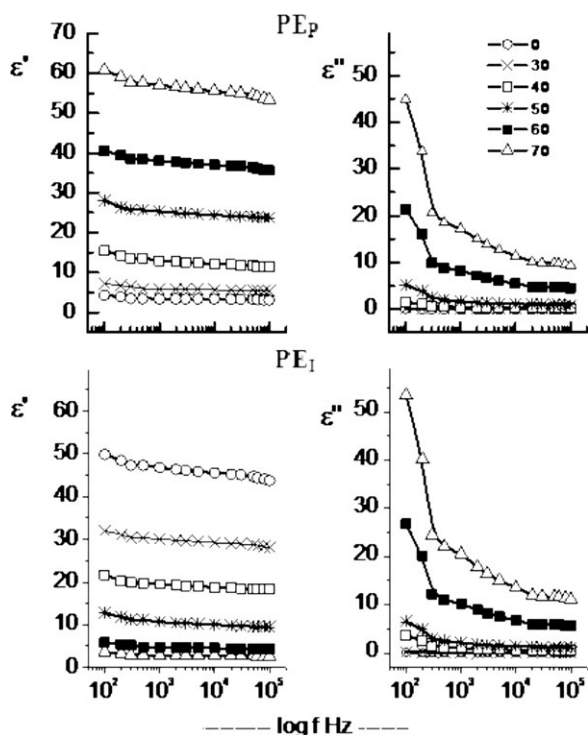


Figure 7. Permittivity ϵ' and dielectric loss ϵ'' versus the applied frequency f , of two styrenated polyester (PE_p and PE_i) and their composites.

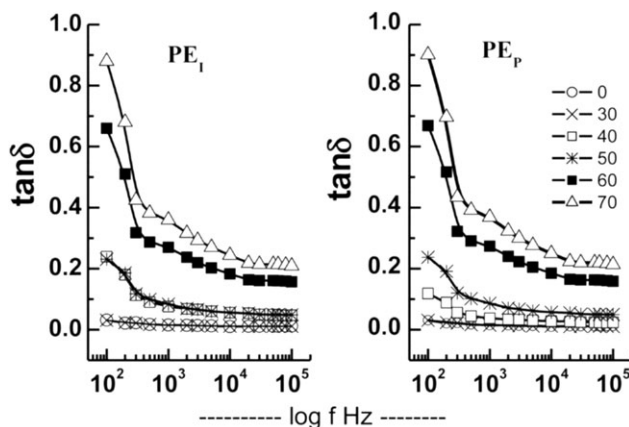


Figure 8. Variation of loss tangent $\tan \delta$ versus the applied frequency of two styrenated polyester (PE_p and PE_i) and their composites.

to a number of degradation reactions leading to the formation of monomer units until the styrenated polyesters are completely volatilize. It was also found that the thermal stability increases with increasing the filler wt%. The thermal stability of polymeric composites was found to be higher than the unfilled polyesters and especially for those of the PE_i.

The initial degradation temperature (T_0), the temperature at 50% weight loss (T_{50}), and the char yield at 600°C are listed in Tables VI and VII. It was noticed that T_0 , T_{50} of carborundum composites increase by increasing the content of filler. The observed improvement in the thermal stability of composite was in terms of delay of the degradation temperatures from 150 to 180°C, and from 150 to 200°C for PE_p and PE_i, respectively. The char yield increases from 2% for the unfilled polyesters to 70% for polyester composite loaded with 70 wt % at 600°C. Also, the char yield of PE_i polymeric composites at higher filler concentrations (50 and 70 wt %) was higher than those of PE_p composites. Increasing the char formation can limit the production of combustible gases, decrease the exothermicity of pyrolysis reaction, decrease the thermal conductivity of the burning materials and restrict their flammability.

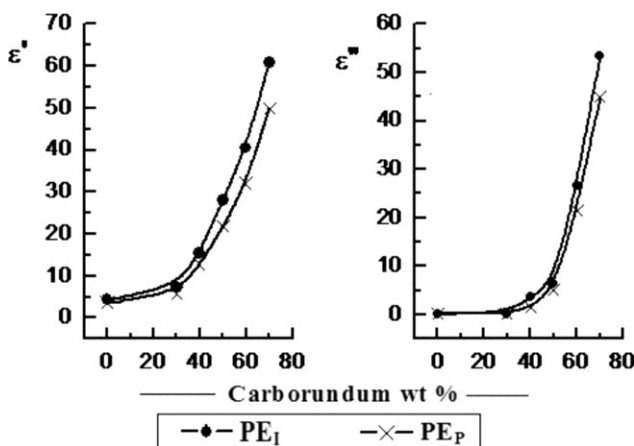


Figure 9. Permittivity ϵ' and dielectric loss ϵ'' at fixed frequency $f = 100$ Hz versus carborundum content of two styrenated polyester (PE_p and PE_i) and their composites.

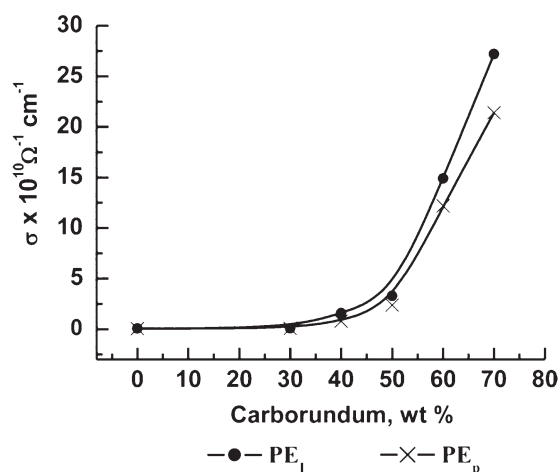


Figure 10. Electrical conductivity σ versus the carborundum contents of two styrenated polyester (PE_p and PE_I) and their composites.

Dielectric Measurements

The permittivity ϵ' and the dielectric loss ϵ'' were measured in the frequency range 100 Hz up to 100 kHz and at room temperature 25°C for two polyesters namely (PE_p and PE_I) filled with carborundum with different concentrations ranging from zero to 70 wt %. The measured data are given in Figure 7 for the two polyesters.

It is clear that ϵ' increases by increasing the percentage of filler, whereas it decreases by increasing the applied frequency which shows anomalous dispersion. The data of ϵ' and ϵ'' given in Figure 7 at different frequencies indicate that an abrupt increase is noticed when the filler concentration increased more than 40 wt %. With this increase, the tendency of conductivity chain formation increase through the aggregation of the filler particles forming network, while with low concentration, the particles of filler are widely dispersed through the polymeric matrix.

Figure 8 illustrates the variation of loss tangent ($\tan \delta$) versus the applied frequency. From this figure, it is clear that $\tan \delta$ increase by increasing filler content but there is no significant change by changing the type of the prepared polyesters. It is noticed that $\tan \delta$ values do not exceed one and that recommends these composites to be used in insulation purposes.

To understand this behavior, the permittivity ϵ' and the dielectric loss ϵ'' were plotted graphically in Figure 9 as a function of filler content at fixed frequency $f = 100$ Hz. It was noticed that after certain concentration (40 wt %) an abrupt increase was occurred. Also, it is interesting to note that both values are slightly higher in case of polyester (PE_I) when they are compared with those obtained for polyester (PE_p). This finding may be due to the fact that the attached group (COOH) in the meta- position in case of PE_I possesses higher polarity than the same group attached in the ortho- position for (PE_p).²⁹

Figure 10 represents the variation of the electrical conductivity σ versus the filler contents for the two investigated polyesters and their composites.

From this figure, it is interesting to find that at the same concentration 40 wt % an abrupt increase was noticed for both

types which confirm the obtained results for ϵ' and ϵ'' . The values of σ are found to be in the order of 10^{-10} which recommend such composites to be used for antistatic applications³⁰ as the electrical conductivity needed for such applications are in the order 10^{-9} to 10^{-14} .

CONCLUSIONS

The presences of isophthalic acid on the backbone structure of polyester affect the properties of polyester and its composites. The polyester based on isophthalic acid (PE_I) imparts highest percent of curing (70/30 wt %) and highest compressive strength values. Also, its composites gave higher compressive strength values and lower water absorption than those based on phthalic anhydride (PE_p). The permittivity ϵ' and the dielectric loss ϵ'' are slightly higher in case of polyester (PE_I) when compared with those obtained for polyester (PE_p).

The incorporation of carborundum filler increase the thermal stability and the char yield of composites than the cured polyesters at high temperature (600°C) and also slightly decrease the water absorption. The physical properties of all composite samples appear to be satisfactorily good. The permittivity ϵ' , the dielectric loss ϵ'' , and electrical conductivity σ of the composites increases by increasing carborundum concentration.

REFERENCES

- Harper, C. A. *Handbook of Plastics and Elastomers*; McGraw-Hill Book Company: New York, 1975.
- Walker, B. M. *Handbook of Thermoplastic Elastomers*; Van Nostrand Reinhold Company: New York, London and Toronto, 1979.
- Czarnomski, T. J. *Unsaturated Polyesters*, *Modern Plastics, Encyclopedia*, 1973, p 66.
- Johnson, K. G.; Yang, L. S. In *Modern Polyesters: Chemistry and Technology of Polymers and Copolyesters*; Scheirs, J., Long, T. E., Eds.; John Wiley: New York, 2003; p 21.
- Mansour, S. H.; Asaad, J. N.; Abd-El-Messieh, S. L. *J. Appl. Polym. Sci.* 2006, 102, 1356.
- Asaad, J. N.; Abd-El-Messieh, S. L. *J. Appl. Polym. Sci.* 2008, 109, 1916.
- Tawfik, S. Y.; Asaad, J. N.; Sabaa, M. W. *Polym. Test.* 2003, 22, 747.
- Composites ASM Handbook/extraction Polyester Resins Prof. H. Hansmann Hochschule Wismar FB MVU, Werkstofftechnologien/Kunststofftechnik Oct. 2003.
- Fink, J. K. *Reactive Polymers Fundamentals and Applications*, William Andrew Publishing: Norwich, NY, 2005; Chapter 1, p 1.
- Malik, M.; Choudhary, V.; Varma, E. K. *J. Macromol. Sci. Rev. Macromol. Chem. Phys.* 2000, C40, 139.
- Salomone, J. C. *Polymeric Materials Encyclopedia*; CRC Press: FL, 1996.
- Lu, M.; Shim, M.; Kim, S. *Eur. Polym. J.* 2001, 37, 1075
- Aziz, S. H.; Ansell, M. P.; Clarke, S. J.; Panteny, S. R. *Compos. Sci. Technol.* 2005, 65, 525.

14. Mouritz, A. P.; Mathys, Z. *Compos. Struct.* **1999**, *47*, 643.
15. Worzakowska, M. *J. Appl. Polym. Sci.* **2009**, *114*, 720.
16. Rother, R. N. *Adv. Polym. Sci.* **1999**, *139*, 67.
17. Tawfik, S.Y.; Asaad, J. N.; Sabaa, M.W. *Polym. Plast. Technol. Eng.* **2004**, *4*, 57.
18. Hristova, J.; Minster, J. *J. Appl. Polym. Sci.* **2003**, *89*, 3329.
19. Charles A. Harper Handbook of Plastics, Elastomers, and Composites, 4th ed.; McGraw-Hill Companies, Inc.: New York, **2002**.
20. Zhu, K.; Schmauder, S. *Comput. Mater. Sci.* **2003**, *28*, 743.
21. Rusu, M.; Sofian, N.; Rusu, D. *Polym. Test.* **2001**, *20*, 409.
22. Patnaik, A.; Satapathy, A.; Mahapatra, S. S.; Dash, R. R. *J. Reinforc. Plast. Compos.* **2008**, *27*, 1093.
23. Abd-El-Messieh, S. L.; Abd-El-Nour, K. N. *J. Appl. Polym. Sci.* **2003**, *88*, 1613.
24. Janković, B. *Chem. Eng. J.* **2010**, *162*, 331.
25. Mansour, S. H.; Abd-El-Messieh, S. L. *J. Appl. Polym. Sci.* **2002**, *83*, 1167.
26. Tagliavia, G.; Porfiri, M.; Gupta, N. *Compos. B: Eng.* **2010**, *41*, 86.
27. Rozman, H. D.; Musa, L.; Azniwati, A. A.; Rozyanty, A. R. *J. Appl. Polym. Sci.* **2011**, *119*, 2549.
28. Ikladious, N. E.; Doss, N. L.; Moussa, H. E.; Nassif, J. *Polym.-Plast. Technol. Eng.* **1997**, *36*, 733.
29. AbdEl-Nour, K. N.; Abd-El-Messieh, S. L.; El-Sabee, M. Z. *Mol. Liq.* **1995**, *63*, 265.
30. Huang, J.-C. *Adv. Polym. Technol.* **2002**, *21*, 299.