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SYNTHESIS AND CHARACTERIZATION OF POLYESTERIMIDES CONTAINING ETHER LINKAGES

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ABSTRACT

Three novel dicarboxylic acids, bis-4,4′-[*N*-4(4′-hydroxycarbonyl phenyleneoxy) phthalimido] diphenyl sulfone, bis-4,4′-[*N*-4(4′-hydroxycarbonyl phenyleneoxy) phthalimido] diphenyl methane, and bis-4,4′-[*N*-4(4′-hydroxycarbonyl phenyleneoxy) phthalimido] diphenyl ether, were synthesized, and several polyesterimides were prepared from diacid chlorides and bisphenols by solution polycondensation. The polymers were obtained in 65–88% yield and had inherent viscosities in the 0.18 to 0.64 dL/g range. The polymers were characterized by IR, elemental analysis, x-ray, TGA, DSC, and solubility tests. All the polymers were readily soluble in polar aprotic solvents and had a 10% weight loss temperature above 375°C in nitrogen.

INTRODUCTION

Wholly aromatic polyimides are thermally the most stable engineering plastics. However, their applications are somewhat limited due to processing difficulties like insolubility in conventional solvents and their extremely high softening or melting temperatures [1–3]. Therefore, much effort has been made to develop structurally such modified aromatic polyimides as polyetherimides [4–6], polyamideimides [7–9], and polyesterimides [10–16] with improved solubility.

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The present paper describes the synthesis and properties of several polyesterimides from three novel dicarboxylic acids (IVa–c) containing preformed imide rings. The main advantage of this approach is that the monomer carries the imide ring and hence these polymers do not require any postcuring at high temperatures. Aromatic ether linkages were incorporated in the polymer-forming diacids in order to increase solubility and hence processability of the polymers.

EXPERIMENTAL

Measurements

IR spectra were obtained on a Perkin-Elmer 781 infrared spectrophotometer, and ^{1}H-NMR spectra were recorded on a Varian EM390 instrument using TMS as the internal standard. X-ray diffractograms were obtained on a Philips PW instrument using nickel-filtered CuK$_\alpha$ radiation. Thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) were performed on a Perkin-Elmer TGS-7 unit at a heating rate of 20°C/min. Elemental analysis was done on a Perkin-Elmer 240. Inherent viscosities were determined at a concentration of 0.5 g/dL in *N,N*-dimethyl acetamide, and densities were determined with a small pycnometer using hexane as a nonsolvent at 30°C.

Monomer Preparation

Commercial hydroquinone, Bisphenol A, 1,5-dihydroxy naphthalene, 4,4′-dihydroxy diphenylsulfide, and 4,4′-dihydroxy diphenylsulfone were recrystallized before use. Chloroform, pyridine, and thionyl chloride were distilled prior to use. All other reagents were of analytical grade.

Bis-4-[*N*-(4-nitrophthalimido)phenyl]sulfone (IIIa), Bis-4-[*N*-(4-nitrophthalimido)phenyl]methane (IIIb), Bis-4-[*N*-(4-nitrophthalimido)phenyl]ether (IIIc)

The bis(4-nitrophthalimides) (IIIa–c) were prepared by the condensation of 4-nitrophthalic anhydride with the corresponding diamine. A typical procedure adopted is as follows.

To a stirred solution of 4,4′-diaminodiphenyl sulfone (6.21 g, 0.025 mol) in DMAc (75 mL) was added 4-nitrophthalic anhydride (9.67 g, 0.0501 mol) and was left stirred at room temperature for 30 minutes. To this, toluene (30 mL) was added and the resulting mixture was heated under reflux for 5 hours while removing water azeotropically. It was then poured into water, and the precipitated dinitro compound (IIIa) was filtered, washed with water, dried, and recrystallized from acetic acid.

Bis-4,4′-[*N*-4(4′-hydroxycarbonyl phenyleneoxy) phthalimido] diphenyl sulfone (IVa), Bis-4,4′-[*N*-4(4′-hydroxycarbonyl phenyleneoxy) phthalimido] diphenyl methane (IVb), Bis-4,4′-[*N*-4(4′-hydroxycarbonyl phenyleneoxy) phthalimido] diphenyl ether (IVc)

A typical procedure used for the preparation of diacids IVa–c is given below.

To a three-necked round-bottom flask equipped with a Dean–Stark trap was added 4-hydroxy benzoic acid (5.53 g, 0.04 mol), potassium hydroxide (4.49 g, 0.08

mol), DMSO (75 mL), and toluene (30 mL). It was then heated to 100°C while removing water azeotropically. After complete dehydration, the excess toluene was removed. The reaction mixture was cooled to 60°C and bis-4-[*N*-(4-nitrophthalimido) phenyl]sulfone (11.97 g, 0.02 mol) and DMSO (25 mL) were added and heated at 100°C for 6 hours under N_2. The cooled reaction mixture was poured into acetone and stirred well. The dipotassium salt obtained was filtered, dissolved in water, and the dicarboxylic acid was precipitated out by the addition of sulfuric acid. The diacid (IVa) was filtered, washed with distilled water, dried, and recrystallized from acetone.

Bis-4,4′-[*N*-4(4′-hydroxycarbonyl phenyleneoxy) phthalimido] diphenyl sulfone diacyl chloride (Va), Bis-4,4′-[*N*-4(4′-hydroxycarbonyl phenyleneoxy) phthalimido] diphenyl methane diacyl chloride (Vb), Bis-4,4′-[*N*-4(4′-hydroxy carbonyl phenyleneoxy) phthalimido] diphenyl ether diacyl chloride (Vc)

The diacid chlorides (Va–c) were prepared by refluxing the corresponding diacids with excess thionyl chloride.

The bis(4-nitrophthalimides) (IIIa–c) and diacids (IVa–c) were characterized by melting points, IR, ^{1}H NMR, and elemental analysis (Table 1). The diacid chlorides, however, were not studied by ^{1}H NMR since they were hygroscopic, poorly soluble in $CDCl_3$, and rapidly reverted to the free acids in DMSO-d_6 as well as trifluoro acetic acid.

MODEL REACTIONS

Preparation of MDE-1

To a mixture of Va (1.20 g, 0.0015 mol), chloroform (20 mL), and pyridine (5 mL), methanol (5 mL) was added and stirred for 12 hours. The solution was then poured into methanol, and the precipitated product was filtered, washed with bicarbonate solution, water, and dried.

Yield: 85%
Mp: 216–17°C
IR (KBr): 2950 cm^{-1} [C—H stretching (—CH_3)]
1775 cm^{-1} [imide carbonyl (symmetric stretching)]
1720 cm^{-1} [ester carbonyl and imide carbonyl(asymmetric stretching)]
1265 cm^{-1} [C—O stretching]
720 cm^{-1} [imide ring]
^{1}H NMR (d_6-DMSO + $CDCl_3$): δ 3.8 (s, 6H, —CH_3)
δ 6.75–8.1 (m, 22H, aromatic protons)
Analysis: Calculated for $C_{44}H_{28}O_{12}N_2S$
Calculated: C, 65.34%; H, 3.49%; N, 3.46%
Found: C, 65.48%; H, 3.40%; N, 3.56%

TABLE 1. Physical Characteristics and Spectral Data of Bis(4-nitrophthalimide)s (IIIa–c), Diacids (IVa–c), and Diacid Chlorides (Va–c)

Code	Yield, %	Melting point, °C	IR (KBr), cm^{-1}	^1H NMR chemical shifts (δ) in DMSO-d_6[a]
IIIa	93	324–26	1775 and 1720 (symmetric and asymmetric stretching of imide carbonyl), 1530 and 1335 ($-NO_2$), 1360 and 1150 ($-SO_2-$), 720 (imide ring)	7.3–8.9 (m, 14H, Ar)
IIIb	96	318–20[b]	1775 and 1720 (symmetric and asymmetric stretching of imide carbonyl), 1530 and 1335 ($-NO_2$), 720 (imide ring)	7.0–8.8 (m, 14H, Ar) 3.8 (s, $-CH_2-$)
IIIc	95	321–22[c]	1775 and 1720 (symmetric and asymmetric stretching of imide carbonyl), 1530 and 1335 ($-NO_2$), 720 (imide ring)	6.8–8.8 (m, 14H, Ar)
IVa	93	281–83	3400 ($-OH$), 1780 and 1720 (imide carbonyl), 1680 (acid carbonyl), 1365 and 1150 ($-SO_2-$), 720 (imide ring)	6.8–8.2 (m, 22H, Ar) 9.8 ($-COO\underline{H}$)
IVb	92	255–56	3400 ($-OH$), 1775 and 1720 (imide carbonyl), 1675 (acid carbonyl), 720 (imide ring)	3.8 (s, $-CH_2-$) 6.7–8.15 (m, 22H, Ar) 9.85 ($-COO\underline{H}$)
IVc	91	228–30	3400 ($-OH$), 1775 and 1715 (imide carbonyl), 1670 (acid carbonyl), 720 (imide ring)	6.8–8.2 (m, 22H, Ar) 9.65 ($-COO\underline{H}$)
Va	97	180	1775 (strong band) [$-\underline{CO}Cl$ and imide carbonyl (symmetric stretching)], 1720 [imide carbonyl asymmetric stretching], 1365 and 1150 ($-SO_2-$), 720 (imide ring)	–
Vb	93	137–39	1780 (strong band) [$-\underline{CO}Cl$ and imide carbonyl (symmetric stretching)], 1720 [imide carbonyl asymmetric stretching], 725 (imide ring)	–
Vc	92	168–70	1775 (strong band) [$-\underline{CO}Cl$ and imide carbonyl (symmetric stretching)], 1720 [imide carbonyl asymmetric stretching], 720 (imide ring)	–

[a] The diacid chlorides were not studied by ^1H NMR since they were hygroscopic, poorly soluble in $CDCl_3$, and rapidly reverted to the free acids in DMSO-d_6 as well as trifluoro acetic acid.

[b] Reference 7: 317–20°C.

[c] Reference 7: 322–23°C; Ar = aromatic, s = singlet, m = multiplet.

Preparation of MDE-2

This was prepared from Vb (1.12 g, 0.0015 mol) and methanol (5 mL) by the same procedure as described for the preparation of MDE-1.

Yield: 82%
Mp: 191°C
IR (KBr): 2950 cm^{-1} [C—H stretching (—CH_3)]
1770 cm^{-1} [imide carbonyl (symmetric stretching)]
1715 cm^{-1} [ester carbonyl and imide carbonyl(asymmetric stretching)]
1260 cm^{-1} [C—O stretching]
720 cm^{-1} [imide ring]
^{1}H NMR (d_6-DMSO + $CDCl_3$): δ 3.8 (s, 2H, —CH_2—)
δ 3.9 (s, 6H, —CH_3)
δ 6.8–8.0 (m, 22H, aromatic protons)
Analysis: Calculated for $C_{45}H_{30}O_{10}N_2$
Calculated: C, 71.24%; H, 3.99%; N, 3.69%
Found: C, 71.13%; H, 4.07%; N, 3.71%

Preparation of MDE-3

This was prepared from Vc (1.13 g, 0.0015 mol) and methanol (5 mL) as per the procedure given for compound MDE-1.

Yield: 81%
Mp: 182–83°C
IR (KBr): 2950 cm^{-1} [C—H stretching (—CH_3)]
1775 cm^{-1} [imide carbonyl (symmetric stretching)]
1720 cm^{-1} [ester carbonyl and imide carbonyl(asymmetric stretching)]
1265 cm^{-1} [C—O stretching]
720 cm^{-1} [imide ring]
^{1}H NMR (d_6-DMSO + $CDCl_3$): δ 3.9 (s, 6H, —CH_3)
δ 6.9–8.15 (m, 22H, aromatic protons)
Analysis: Calculated for $C_{44}H_{28}O_{11}N_2$
Calculated: C, 69.47%; H, 3.71%; N, 3.68%
Found: C, 69.34%; H, 3.76%; N, 3.60%

POLYMERIZATION

A typical polymerization procedure is given below.

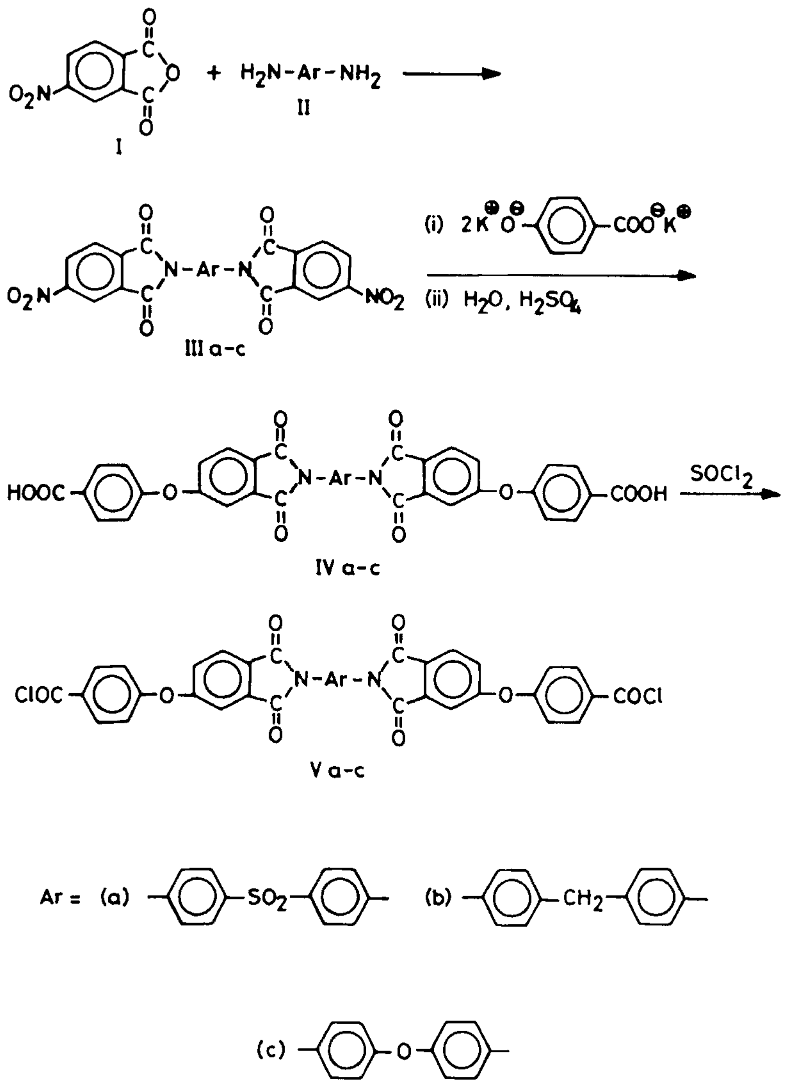
Preparation of PEI-1

To a stirred solution of Va (1.20 g, 0.0015 mol) in chloroform (20 mL) and pyridine (5 mL), Bisphenol A (0.34 g, 0.0015 mol) was added and stirred for 12 hours. The reaction mixture was poured into methanol (200 mL) and the precipitated polymer was filtered, washed with dilute sodium bicarbonate solution, water, and dried.

RESULTS AND DISCUSSION

Bis-4-[*N*-(4-nitrophthalimides)] (IIIa–c) were prepared by the condensation of 4-nitrophthalic anhydride with the corresponding diamine in *N,N*-dimethyl acetamide. The complete cyclization of the intermediate amic-acid was achieved by toluene–water azeotropic distillation. The IR, NMR, and elemental analysis of IIIa–c are in accord with the proposed structures (Table 1).

The diacids IVa–c were prepared by the nucleophilic displacement of nitro groups from the corresponding bis(4-nitrophthalimides) (IIIa–c) (Scheme 1). The displacement of nitro groups from IIIa–c was facile due to activation by the cyclic



SCHEME 1.

imide group, and it thereby led to pure diacids (IVa–c) in high yields. The IR (Fig. 1), NMR, and elemental analysis of the diacids agreed well with the proposed structures (Table 1). The diacyl chlorides (Va–c) were prepared by refluxing the corresponding diacid with thionyl chloride. The diacid chlorides were characterized by IR and melting points (Table 1).

To characterize the diacids IVa–c and to help in structural identification of polymers, model compounds (MDE-1, MDE-2, and MDE-3) were prepared by treating the corresponding diacid chloride with an excess of methanol in the presence of pyridine. The ^{1}H-NMR spectrum of MDE-1 showed a singlet at δ 3.80, while MDE-2 and MDE-3 showed a singlet at δ 3.90 due to the methyl protons ($-COO\underline{CH_3}$) (Table 1). The elemental analyses of the model compounds were in accord with the proposed structures. Several polyesterimides (PEI-1 to PEI-15) were prepared using the diacyl chlorides, Va–c, and bisphenols (Scheme 2). The polymerization reaction was carried out in chloroform-pyridine (4:1 v/v) mixture at room temperature for 12 hours. The results of polymerization such as yields, densities, etc. are summarized in Table 2.

IR spectra of model compounds showed absorption bands at 2950 cm^{-1} (small absorption band) [C—H stretching of $-CH_3$]; 1775 cm^{-1} (MDE-1), 1770 cm^{-1} (MDE-2), 1775 cm^{-1} (MDE-3) [imide carbonyl (symmetric stretching)]; 1720 cm^{-1} (MDE-1), 1715 cm^{-1} (MDE-2), 1720 cm^{-1} (MDE-3) [ester carbonyl and imide carbonyl (asymmetric stretching)], and 1265 cm^{-1} (MDE-1), 1260 cm^{-1} (MDE-2), 1265 cm^{-1} (MDE-3) [C—O stretching]. The 1710–1720 cm^{-1} region should represent two

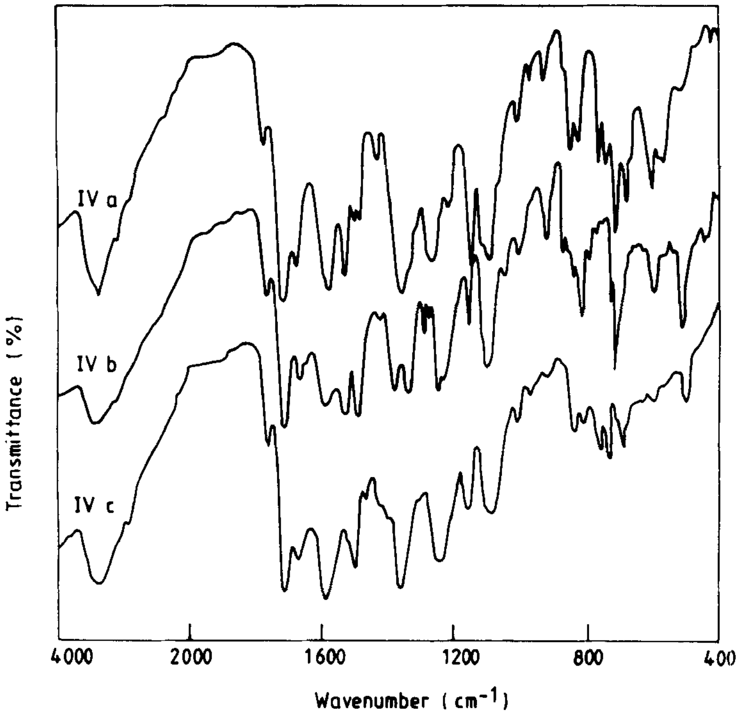
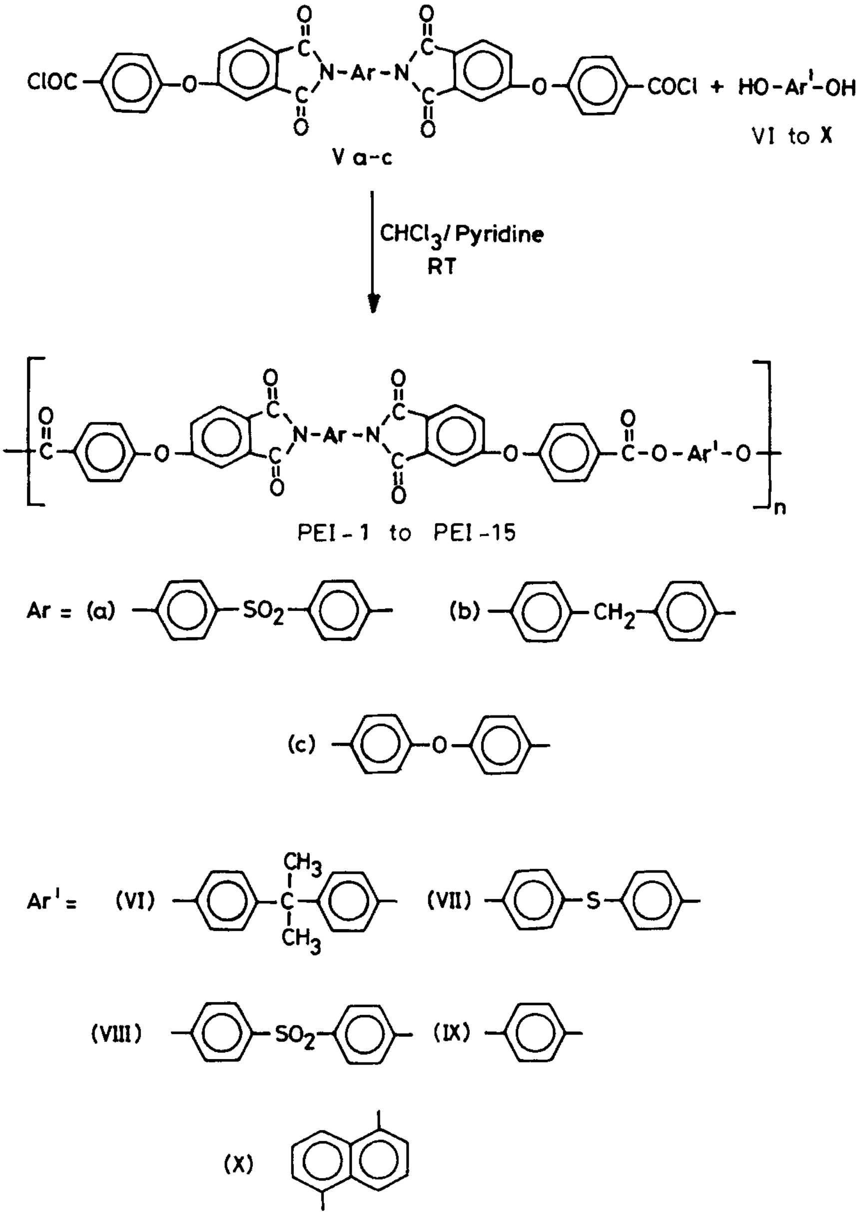


FIG. 1. IR spectra of diacids (IVa, IVb, IVc).



SCHEME 2.

carbonyl peaks due to imide carbonyl (asymmetric stretching) and ester carbonyl. However, the two carbonyls individually are so strong that they become inseparable, and the overall absorption is seen as a very strong and broad band centering around 1720 cm^{-1}. This is not surprising since two other literature citations [13, 14] on polyesterimides have expressed a similar difficulty. The IR spectra of the polymers showed absorption bands at 1775 ± 5 cm^{-1} [imide carbonyl (symmetric stretch-

TABLE 2. Preparation of Polyesterimides

Polymer code	Diacid chloride	Bisphenol	Yield, %	η_{inh},[a] dL/g	Density,[b] g/cm^3
PEI-1	Va	VI	75	0.52	1.06
PEI-2	Va	VII	68	0.47	1.12
PEI-3	Va	VIII	71	0.36	1.08
PEI-4	Va	IX	85	0.25	1.20
PEI-5	Va	X	88	0.18	1.29
PEI-6	Vb	VI	70	0.55	1.01
PEI-7	Vb	VII	71	0.40	1.15
PEI-8	Vb	VIII	73	0.38	1.18
PEI-9	Vb	IX	82	0.26	1.24
PEI-10	Vb	X	88	0.21	1.20
PEI-11	Vc	VI	66	0.64	0.96
PEI-12	Vc	VII	65	0.56	1.04
PEI-13	Vc	VIII	72	0.45	0.98
PEI-14	Vc	IX	76	0.32	1.16
PEI-15	Vc	X	83	0.26	1.11

[a] Measured in DMAc at 30°C, $C = 0.2$ g/dL.
[b] Determined pynometrically in hexane at 30°C.

TABLE 3. Solubilities of Polyesterimides[a]

Polymer code	NMP, DMAc	DMF	*m*-Cresol	Pyridine	Chloroform	Benzene, acetone	Conc H_2SO_4
PEI-1	++	++	++	++	±	–	++
PEI-2	++	++	±	±	●	–	++
PEI-3	++	++	++	++	–	–	++
PEI-4	++	●	–	●	–	–	++
PEI-5	++	±	–	–	–	–	++
PEI-6	++	++	++	++	±	–	++
PEI-7	++	++	●	++	±	–	++
PEI-8	++	++	++	++	±	–	++
PEI-9	++	++	±	±	–	–	++
PEI-10	++	–	–	–	–	–	++
PEI-11	++	++	++	++	±	–	++
PEI-12	++	++	++	++	±	–	++
PEI-13	++	++	++	++	●	–	++
PEI-14	++	++	±	±	–	–	++
PEI-15	++	++	±	±	–	–	++

[a] ++ = Soluble at room temperature. ± = Soluble on heating. – = Insoluble.

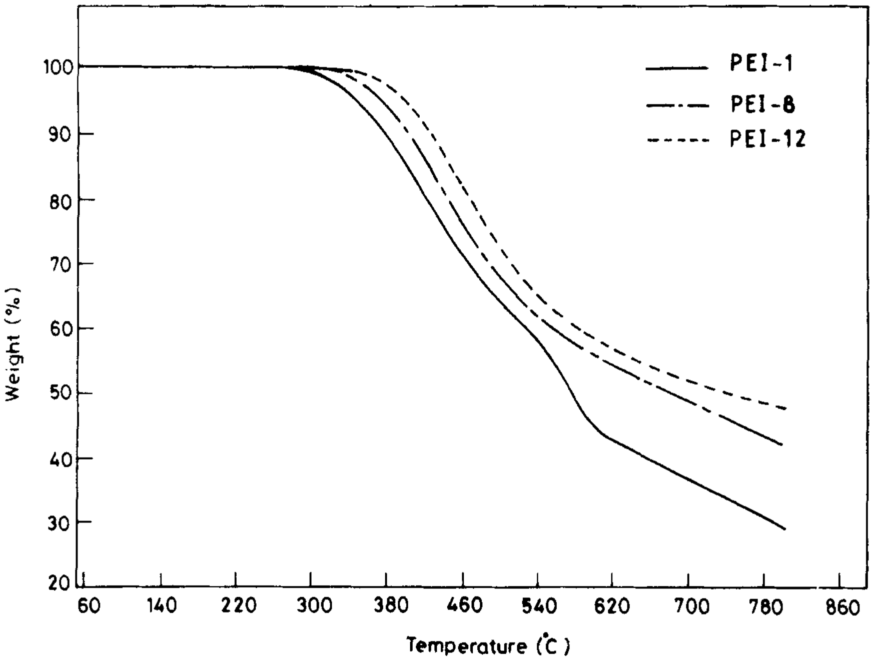


FIG. 2. TGA curves of polyesterimides (PEI-1, PEI-8, PEI-12) in nitrogen.

ing)], 1715 ± 5 cm^{-1} [ester carbonyl and imide carbonyl (asymmetric stretching)], and 1250 ± 15 [C—O stretching]. The strong absorption band of ester carbonyl overlaps with the strong absorption band of imide carbonyl at 1715 cm^{-1}. One would expect the higher frequency side of the absorption band at 1710–1720 cm^{-1} to be due to the imide carbonyl (asymmetric stretching) and lower frequency side to the ester carbonyl, with some amount of fine splitting. Unfortunately, such a splitting is not seen in spite of our attempt to zoom and define the carbonyl region with PE-1760 FTIR using a PE-9900 computer. Owing to this difficulty of complete identification of polyesterimide by the IR spectrum, the structures were reestablished by hydrolysis of the polyesterimides, followed by separation and identification of the hydrolyzed products. The elemental analyses of the polyesterimides were in good agreement with the proposed structures.

The inherent viscosities of the polymers measured in *N*,*N*-dimethylacetamide at 30°C were in the 0.18 to 0.64 dL/g range. The polymers derived from hydroquinone and 1,5-dihydroxy naphthalene were found to have lower viscosities, probably due to the poor solubility of the growing polymer chains in the reaction medium.

Solubility Characteristics

A 10% solution was taken as a criterion for solubility. All these polymers were soluble in *N*-methyl-2-pyrrolidone (NMP), *N*,*N*-dimethylacetamide (DMAc), *N*,*N*-dimethylformamide (DMF) and concentrated H_2SO_4 (Table 3). It is noteworthy that some of the polymers were completely or partially soluble even in *m*-cresol and pyridine. The improved solubilities of the polymers over the well-documented

TABLE 4. Thermal Properties of Polyesterimides[a]

	TGA					DSC	
	Temperature at various percentage decompositions, °C				Char yield,[b] %	T_g,[c] °C	T_{exo},[d] °C
Polymer code	5	10	20	30			
PEI-1	351	379	425	467	30	196	548
PEI-2	400	428	473	509	43	210	618
PEI-3	369	396	457	497	45	224	585
PEI-4	420	449	490	534	53	247	608
PEI-5	432	462	508	559	60	262	583
PEI-6	364	390	420	448	39	189	488
PEI-7	383	411	451	518	48	202	615
PEI-8	372	399	438	485	43	–	568
PEI-9	398	421	471	518	50	230	604
PEI-10	413	440	491	563	60	246	596
PEI-11	362	386	425	460	37	180	552
PEI-12	397	431	467	512	48	196	620
PEI-13	375	403	441	489	50	204	567
PEI-14	430	462	502	548	51	223	608
PEI-15	442	472	517	574	60	235	602

[a] Heating rate 20°C/min.

[b] Percent char yield at 800°C.

[c] Temperature as observed in the first heating traces of DSC thermogram.

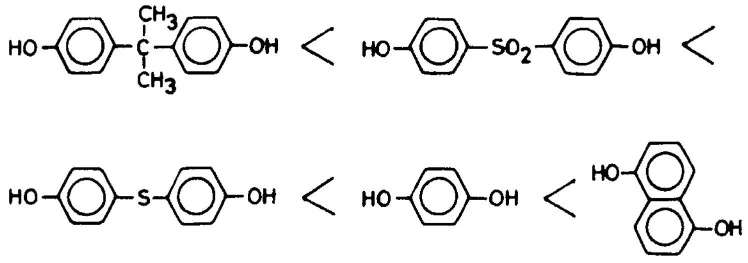
[d] Temperature at which maximum exothermal peak observed.

polyesterimides [12, 14] may be explained by the fact that the incorporation of aromatic ether linkages into the polymer backbone decreases chain symmetry and, consequently, the degree of crystallinity, which in turn increases solubility.

Thermal Properties

The TGA curves (Fig. 2) of these polymers in N_2 showed rapid weight loss in the 400 to 550°C temperature range. The temperature at which 10% weight loss was observed for these polymers ranged from 379 to 472°C (Table 4). The polymers derived from the diacid IVc showed better thermal stabilities (10% weight loss temperature) than the polymers derived from the diacids IVa and IVb. At 800°C, all the polymers retained more than 40% of their initial weight.

The bisphenols can be arranged in the following order of increasing thermal stability of the polyesterimides.



DSC thermograms of these polymers showed a broad exothermal peak owing to degradation of polymers in the 525 to 650°C temperature range. Endotherms corresponding to the crystalline melt temperature (T_m) were not observed for any of the polymers, while endothermic baseline shifts corresponding to the glass transition temperature (T_g) were observed in the 180 to 258°C temperature range (Table 4). The higher chain stiffness of 1,5-naphthalene and 1,4-phenylene units resulted in relatively higher T_gs of the polyesterimides derived from 1,5-dihydroxy naphthalene and hydroquinone. The polymers derived from IVc had relatively low T_g values as compared to polymers derived from IVa and IVb. The range of T_gs of polyimides is

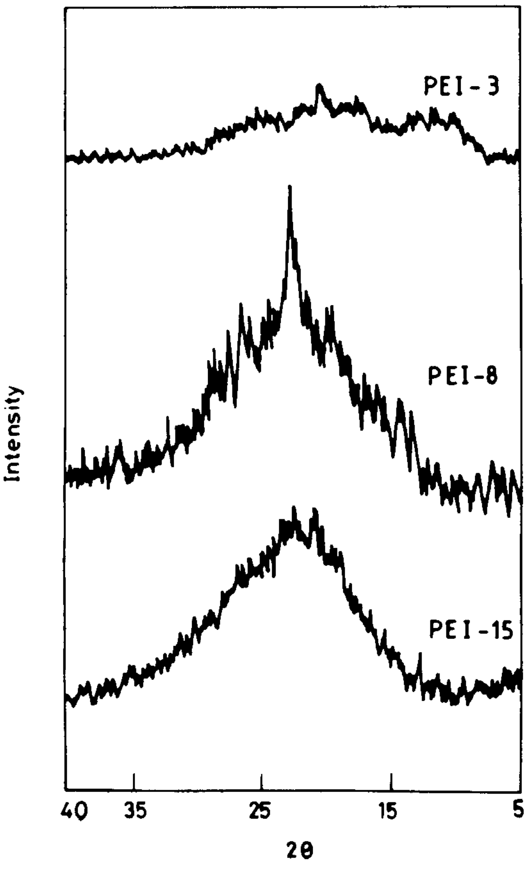


FIG. 3. X-ray diffractograms of polyesterimides (PEI-3, PEI-8, PEI-15).

is very broad, i.e., 200–450°C, and strongly depends on the structure [3]. Incorporation of flexible ether linkages into the polymer backbone increases the freedom of chain motion and decreases T_g. Maiti et al. [13] prepared polyesterimides with an aliphatic backbone and reported T_g in the 180 to 280°C range, while Sato et al. [14] did not observe any T_g for the phenoxaphosphine-containing polyesterimides. The range of T_g (180–258°C) reported in this paper is therefore quite logical, since the variations observed among polyesters [17], polyesterimides [13, 14], and polyimides [13] are found to increase in that order.

Crystallinity

The x-ray diffractograms (Fig. 3) showed that most of these polymers were amorphous in nature. However, the polymers PEI-5, PEI-8, and PEI-14 were partially crystalline.

CONCLUSIONS

These polyesterimides exhibited excellent solubility characteristics with good thermal stabilities. Thus, an increase in solubility and an improvement in processability resulted from the incorporation of flexible aromatic ether linkages.

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