

Facile Synthesis of Novel Optically Active Poly(amide-imide)s Containing *N,N'*-(Pyromellitoyl)-bis-*L*-phenylalanine Diacid Chloride and 5,5-Disubstituted Hydantoin Derivatives Under Microwave Irradiation

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ABSTRACT: Pyromellitic dianhydride (1,2,4,5-benzenetetracarboxylic acid 1,2,4,5-dianhydride) (**1**) was reacted with *L*-phenylalanine (**2**) in a mixture of acetic acid and pyridine (3 : 2) at room temperature, then was refluxed at 90–100°C and *N,N'*-(Pyromellitoyl)-bis-*L*-phenylalanine diacid (**3**) was obtained in quantitative yield. The imide-acid (**3**) was converted to *N,N'*-(Pyromellitoyl)-bis-*L*-phenylalanine diacid chloride (**4**) by reaction with thionyl chloride. Rapid and highly efficient synthesis of poly(amide-imide)s (**6a–f**) was achieved under microwave irradiation by using a domestic microwave oven from the polycondensation reactions of *N,N'*-(Pyromellitoyl)-bis-*L*-phenylalanine diacid chloride (**4**) with six different derivatives of 5,5-disubstituted hydantoin compounds (**5a–f**) in the presence of a small amount of a polar organic medium that acts as a primary microwave

absorber. Suitable organic media was *o*-cresol. The polycondensation proceeded rapidly, compared with the conventional melt polycondensation and solution polycondensation, and was almost completed within 10 min, giving a series of poly(amide-imide)s with inherent viscosities about 0.28–0.44 dL/g. The resulting poly(amide-imide)s were obtained in high yield and are optically active and thermally stable. All of the above compounds were fully characterized by means of FTIR spectroscopy, elemental analyses, inherent viscosity (η_{inh}), solubility test and specific rotation. Thermal properties of the poly(amide-imide)s were investigated using thermal gravimetric analysis (TGA). © 2003 Wiley Periodicals, Inc. *J Appl Polym Sci* 91: 516–524, 2004

Key words: polyamides; polyimides; irradiation

INTRODUCTION

There is a growing interest in poly(amide-imide)s (PAIs) for a variety of applications as they retain good mechanical properties at high temperatures and show easier processability when compared with other aromatic thermostable polymers such as polyamides and polyimides.¹ They are finding numerous applications in adhesives, electronic wire enamel, injection-molding, extrusion products and membranes.^{2–5}

Recently, there has been growing interest in applying microwave energy to synthetic organic chemistry,^{6–9} sample preparation for analysis,¹⁰ extraction of natural products from plants,¹¹ waste treatment,¹² and polymer synthesis.^{13,14} The reactions are very fast and are completed within a short period time. Recently,

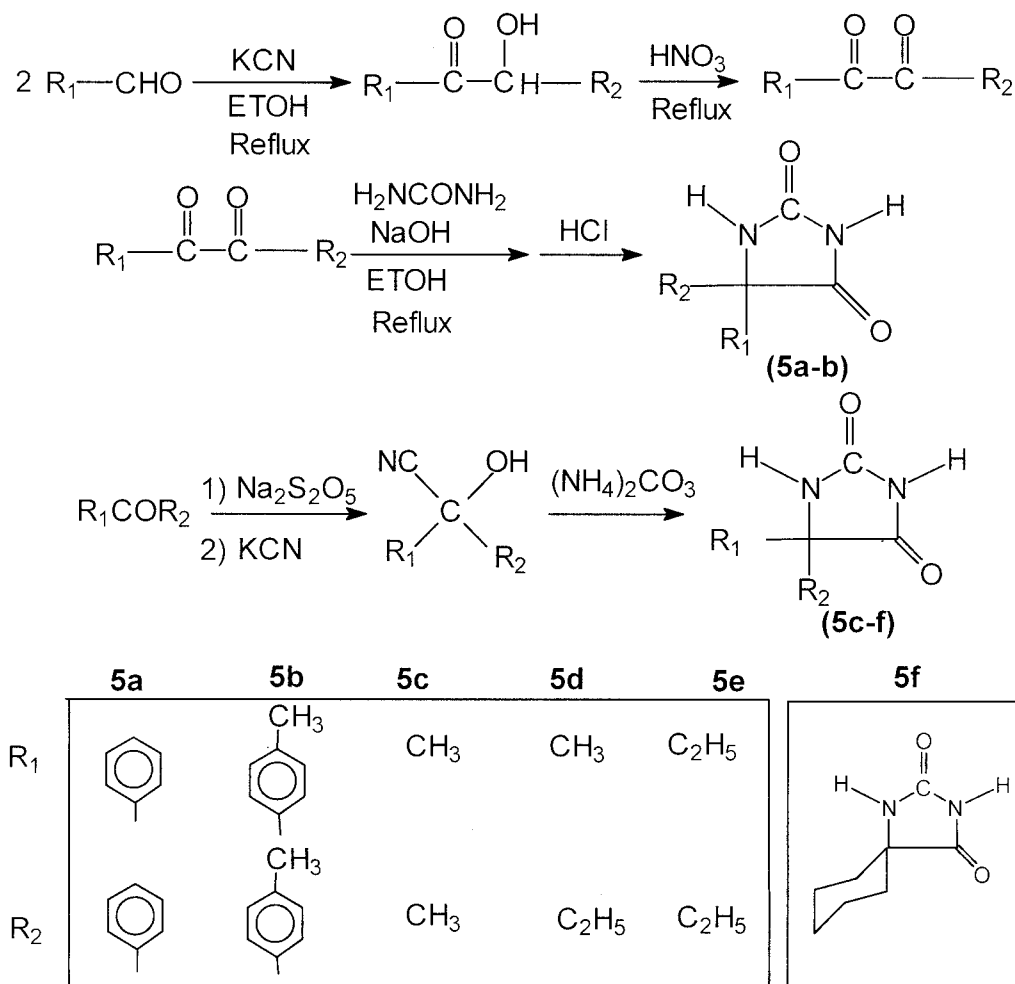
we have used microwave irradiation for the synthesis of poly(amide-imide)s and poly(ester-imide)s.^{15–27}

The synthesis and application of optically active polymers are the newly considerable topics, which have been paid more attention recently, because polymers with chiral structures are biologically very important. Recently, we have synthesized optically active polymers by different methods.^{15–40}

In a previous article,¹⁸ we described a series of novel optically PAIs containing hydantoin and thiohydantoin derivatives moieties as well as two groups of chiral *L*-alanine by the solution polycondensation reactions of 4,4'-carbonyl-bis(phthaloyl-*L*-alanine) diacid chloride as a chiral monomer with six different hydantoin and thiohydantoin derivatives as a heterocycles unit under microwave irradiation. This article describes the synthesis and properties of new optically active PAIs (**6a–f**) from the polycondensation reactions of *N,N'*-(pyromellitoyl)-bis-*L*-phenylalanine diacid chloride (**4**) with six different derivatives of 5,5-disubstituted hydantoin compounds (**5a–f**) as a heterocycles unit under microwave irradiation. Hydantoin and hydantoin derivatives are important intermedi-

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Scheme 1

ates in the synthesis of several amino acids. In the chemical industry various 5,5-disubstituted hydantoin derivatives are the basic of new generation of weatherproof high-temperature-stable epoxy resins.⁴¹

EXPERIMENTAL

Apparatus

A Samsung domestic microwave oven (2450 MHz, 900W) was used for carrying out polycondensation reactions.

Materials

All chemicals were purchased from Fluka Chemical Co. (Switzerland), Aldrich Chemical Co. (Milwaukee, WI), and Merck Chemical Co. (Germany).

Techniques

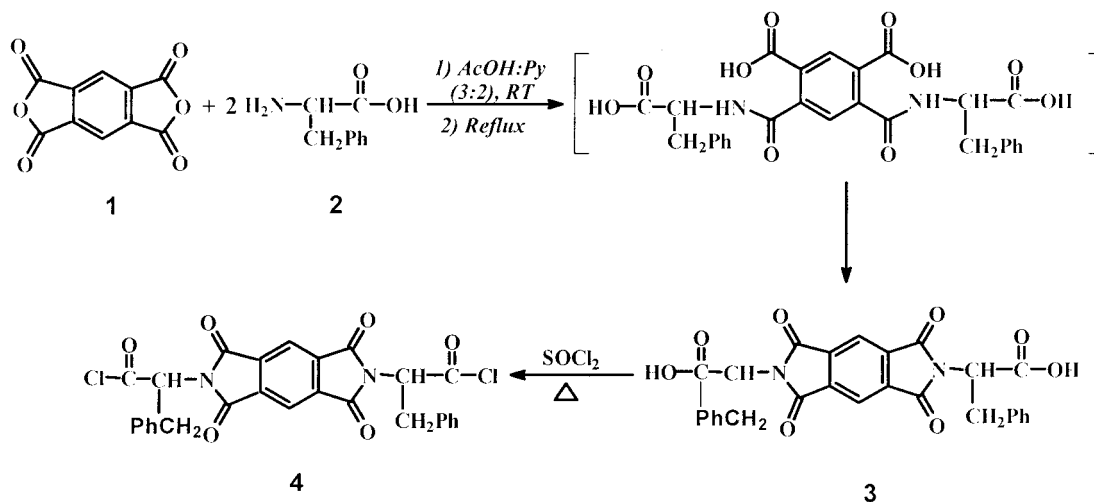
¹H NMR spectra was recorded on a Bruker 500 MHz instrument. Fourier transform infrared (FTIR) spectra

were recorded on Galaxy series FTIR 5000 spectrophotometer (England). Spectra of solids were performed using KBr pellets. Vibrational transition frequencies are reported in wave number (cm⁻¹). Band intensities are assigned as weak (w), medium (m), shoulder (sh), strong (s), and broad (br). Inherent viscosities were measured by a standard procedure using a Technico Regd Trad Merk Viscometer. Specific Rotations were measured by an A-Kruss polarimeter. Thermal Gravimetric Analysis (TGA) data for polymers were taken on a Mettler TA4000 System under N₂ atmosphere at a rate of 10°C/min. Elemental analyses were performed by the Research Institute of Petroleum Industry, Tehran, I.R. Iran. As the source of microwave irradiation, we used a Samsung domestic microwave oven (2450 MHz, 900 W) for carrying out polycondensation reactions.

Monomer synthesis

5,5-disubstituted hydantoin derivative (5a-f)

These compounds were prepared according to a typical procedure that was shown in Scheme 1.¹⁸



Scheme 2

N,N'-(Pyromellitoyl)-bis-L-phenylalanine diacid (3)

Into a 250-mL round-bottomed flask (4.36 g, 20 mmol) of Pyromellitic dianhydride (1,2,4,5-benzenetetracarboxylic acid 1,2,4,5-dianhydride) (1), (6.61 g, 20 mmol) of L-Phenylalanine 2, 80 mL of mixture of acetic acid and pyridine (3 : 2) and a stirring bar were placed. The mixture was stirred at room temperature for overnight and was refluxed for 4 h. The solvent was removed under reduced pressure and the residue was dissolved in 100 mL of cold water, then 5 mL of concentrated HCl was added. The solution was stirred until a white precipitate was formed, then the precipitate filtered off and dried, to give 7.17 g (70%) of compound (3): m.p. 295–298°C, $[\alpha]_D^{25} = +0.2$ (0.05 g in 10 mL DMF); FTIR (KBr): 3500–2800 (m,br), 1776 (sh), 1740–1700 (s, br), 1383 (s), 1363 (s, sh), 1278 (m, br), 1101 (s), 941 (w), 883 (w), 827 (m), 756 (m), 727 (m) cm⁻¹. ¹H NMR (DMSO-*d*₆, TMS): δ 3.29–3.34 (dd, 2H); 3.49–3.53 (dd, 2H); 5.16–5.20 (dd, 2H); 7.09–7.11 (m, 2H); 7.15–7.16 (d, 8H); 8.20–8.21 (s, 2H); 8.57–8.58 (br, 2H) ppm. Analysis: calculated for C₂₈H₂₀N₂O₈: C, 65.61; H, 3.93; N, 5.46; found: C, 65.80; H, 4.20; N, 5.20.

N,N'-(Pyromellitoyl)-bis-L-phenylalanine DIACID chloride (4)

Into a 25-mL round-bottomed flask were placed (2 g, 3.9×10^{-3} mol) of compound 4 and 15 mL of thionyl chloride. The mixture was heated on an oil bath up to 60°C, until the suspension mixture was converted to a clear solution. Then, the solution was stirred overnight at room temperature. Unreacted thionyl chloride was removed under reduced pressure and the residue was washed with dry *n*-hexane two times, to leave 2.04 g (95%) of white crystals. m.p. 189–191°C, $[\alpha]_D^{25} = +0.2$ (0.05 g in 10 mL DMF); FTIR (KBr): 3032 (w), 2916 (w), 1803 (s), 1776 (s), 1740–1700 (s, br), 1496 (w),

1456 (m), 1383 (s, br), 1369 (s, br), 1336 (w), 1107 (m), 937 (w), 844 (m), 734 (s), 528 (m) cm⁻¹. ¹H NMR (500 MHz, CDCl₃, TMS): δ 3.56–3.61 (dd, 2H); 3.69–3.74 (dd, 2H); 5.36–5.45 (dd, 2H); 7.13–7.24 (m, 10H); 8.23 (s, 2H) ppm. Analysis: calculated for C₂₈H₁₈N₂O₆Cl₂: C, 61.21; H, 3.30; N, 5.10; found: C, 61.50; H, 3.50; N, 4.90.

Polymer synthesis

An equimolar mixture of diacid chloride (5) (0.4 g, 7.28×10^{-4} mol) and 5,5-dimethylhydantoin 5a (0.093 g, 7.28×10^{-4} mol) were placed in a reaction vessel and the mixture is ground until fine powder is formed. Then, 0.9 mL of *o*-cresol is added to the mixture and was mixed up until a homogeneous solution is formed. Then the reaction mixture is irradiated in a microwave oven at full power for 10 min. The reaction mixture was poured into 50 mL of methanol. The resulting polymer was filtered off, and was dried, to yield 0.41 g (93%) of brown solid.

All of the other PAIs (6a–f) were synthesized by the above method.

Polymer 6a

FTIR (KBr): 2958 (m), 1774 (s), 1724 (s, br), 1491 (w), 1383 (s, br), 1369 (s), 1221 (m), 1170 (s), 1109 (m), 752 (w), 729 (m) cm⁻¹.

Polymer 6b

FTIR (KBr): 1763 (s), 1724 (s, br), 1458 (w), 1384 (s, br), 1369 (s), 1222 (m), 1170 (m), 1111 (w), 1078 (w) 729 (m) cm⁻¹.

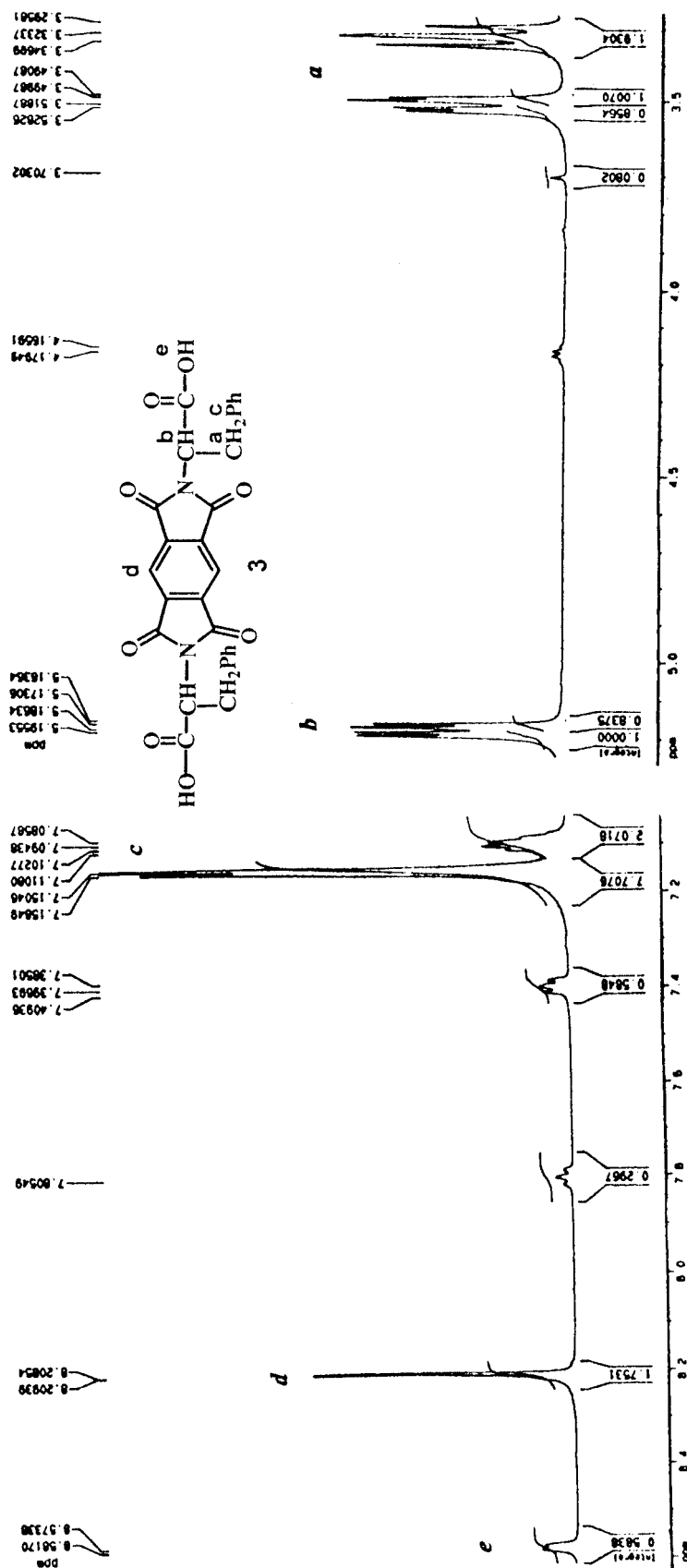


Figure 1 ¹H NMR Spectrum of *N,N'*-(pyromellitoyl)-bis-L-phenylalanine diacid (3).

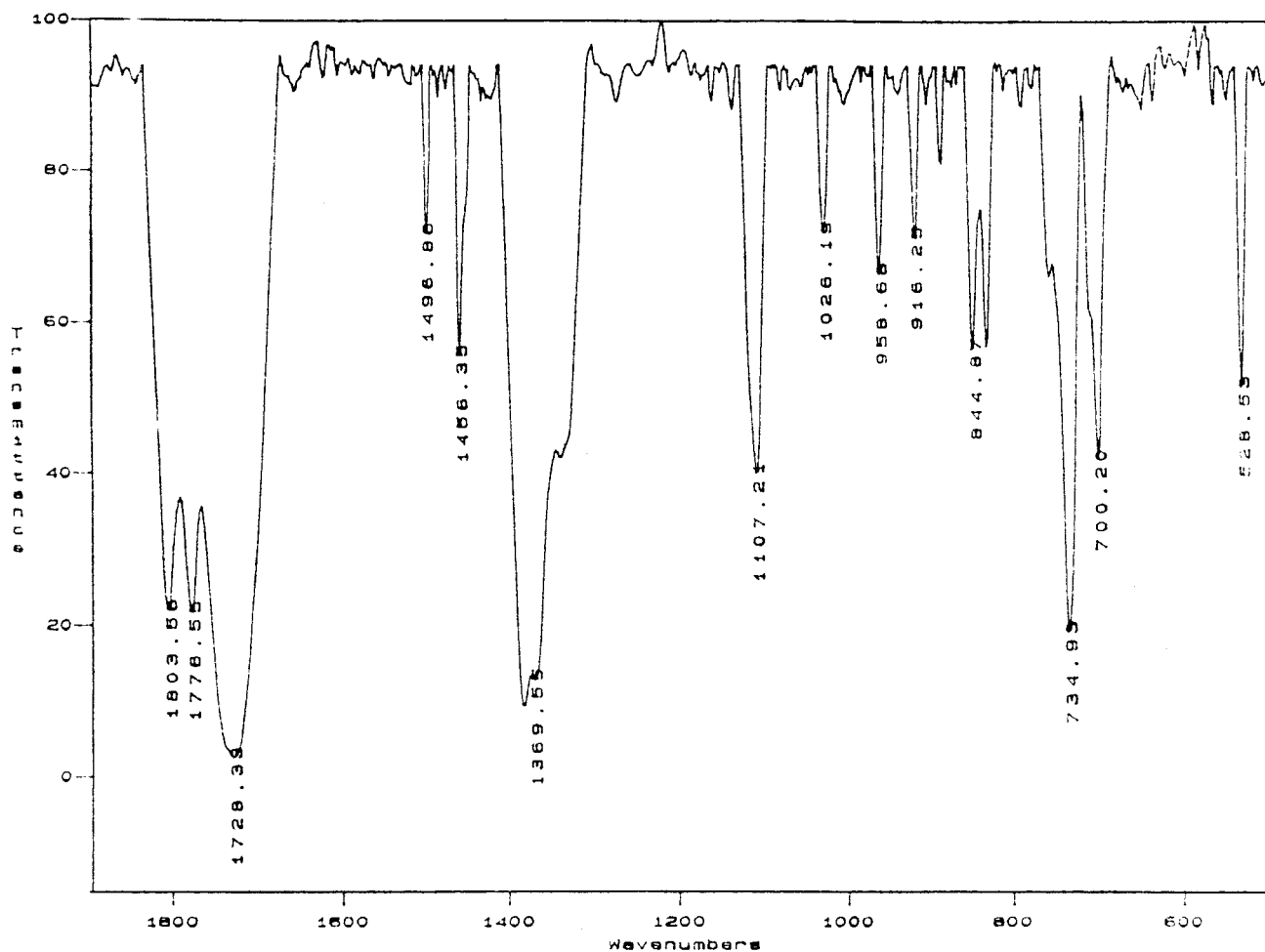


Figure 2 FTIR spectrum of *N,N'*-(pyromellitoyl)-bis-*L*-phenylalanine diacid chloride (4).

Polymer 6c

FTIR (KBr): 1764 (s), 1724 (s, br), 1491 (m), 1458 (m), 1384 (s, br), 1369 (s), 1222 (s), 1170 (s), 1109 (m), 1074 (m), 1020 (w), 841 (w), 752 (m), 729 (m) cm^{-1} .

Polymer 6d

FTIR (KBr): 1764 (s), 1726 (s, br), 1491 (m), 1460 (m), 1383 (s), 1369 (s, sh), 1222 (s), 1170 (s), 1108 (m), 1076 (m), 1020 (w), 841 (m), 752 (m), 729 (w) cm^{-1} .

Polymer 6e

FTIR (KBr): 1764 (s), 1724 (s,br), 1491 (w), 1458 (w), 1384 (s), 1369 (s, sh), 1222 (s), 1170 (s), 1111 (m), 1076 (m), 1020 (w), 839 (w), 752 (w), 729 (m) cm^{-1} .

Polymer 6f

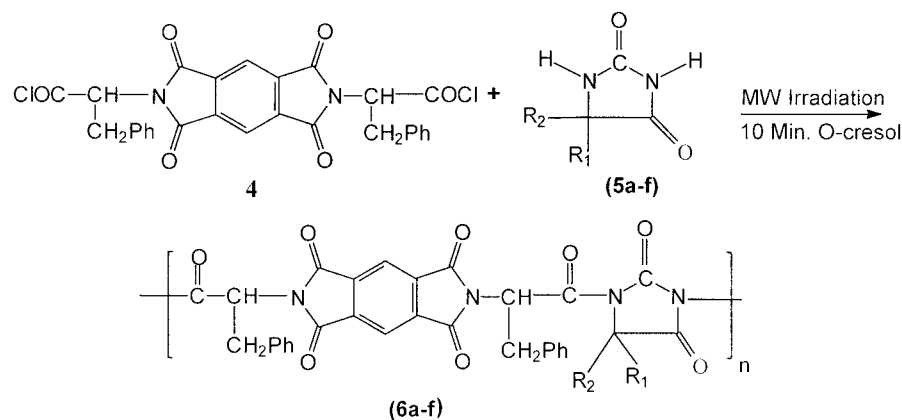
FTIR (KBr): 1767 (s), 1724 (s, br), 1491 (w), 1458 (w), 1384 (s, br), 1369 (s, br), 1222 (m), 1170 (m), 1111 (m), 1070 (m), 1020 (w), 840 (w), 752 (w), 735 (m) cm^{-1} .

RESULTS AND DISCUSSION

Monomer synthesis

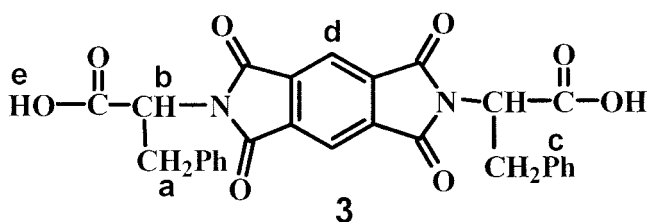
N,N'-(Pyromellitoyl)-bis-*L*-phenylalanine diacid chloride (4) was prepared in two steps as shown in Scheme 2.

The asymmetric diacid (3) was synthesized by the condensation reaction of two equimolar of *L*-phenylalanine with one equimolar of dianhydride (1) in a mixture of acetic acid–pyridine (3 : 2). Dissolving the residue in cold water gives a gummy like solid that breaks by adding concentrated HCl and gave a white color solid. The resulting asymmetric diacid (3) was converted to its diacid chloride derivative (4) by the reaction with thionyl chloride. The monomer (4) was purified by washing with *n*-hexane. The chemical structure and purity of the optically active monomers (3) and (4) were proved using elemental analysis, FTIR, and ^1H NMR spectroscopic techniques. The ^1H NMR spectrum of compound (3) showed peaks between 3.29–3.53 ppm as a doublet of doublet that were assigned to the benzylic protons (a). Peaks between



Scheme 3

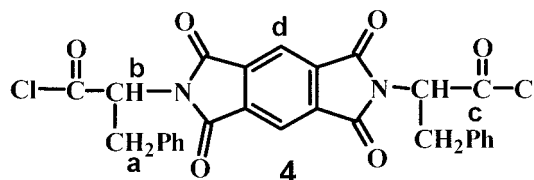
5.16–5.20 ppm as a doublet of doublet which were assigned to the CH(b) protons, which is a chiral center. The peaks between 7.09–7.16 ppm were assigned to aromatic protons (c), peaks between at 8.20–8.21 ppm were assigned to pyrimillitimide ring protons (d) and peaks between at 8.55 ppm was assigned to COOH (e) proton (Fig. 1).



The FTIR spectrum of compound (3) showed a broad and strong peak between 2800–3500 cm^{-1} , which was assigned to the COOH groups and three absorption bands at 1780 and 1700 cm^{-1} , which are characteristic peaks for imide rings and acidic groups. Disappearance of strong acidic hydroxyl peak in FTIR spectrum of compound (4) confirmed a complete conversion of diacid (3) to diacid chloride (4). On the other hand, because of the electron-withdrawing character of the Cl group, the three carbonyl peak of diacid chloride in

comparison with its starting diacid, were shifted to higher frequency (Fig. 2).

The ^1H NMR spectrum of compound (4) showed a doublet of doublet peaks between 3.56–3.74 ppm, which were assigned to the benzylic protons (a). Peaks between 5.36–5.45 ppm a doublet of doublet that were assigned to the CH(b) protons, which is a chiral center. The peak between 7.13–7.24 ppm were assigned to aromatic protons (c) and peak at 8.23 ppm was assigned to pyrimillitimide ring protons (d).



Polymer synthesis

PAIs (6a–f) were synthesized by microwave-assisted polycondensation reactions of an equimolar mixture of monomer (4) with six different derivatives of 5,5-disubstituted hydantoin compounds (5a–f) in *o*-cresol as organic solvent by using a domestic microwave oven as shown in Scheme 3. The *o*-cresol was used as polar organic medium that acts as a primary microwave absorber and as a solvent for both of the starting monomers and the resulting polymers, which allows induction of effective homogeneous heating of the reaction mixture and thereby subsequent polycondensation reaction leading to the polymer formation. The optimum period of reaction time (irradiation time) was found to be 10 min, and below this time will give polymers with lower inherent viscosities and above this time the materials will degradate.

Polymer characterization

Synthesis and some physical properties of PAIs (6a–f) are summarized in Table I. These polymers have in-

TABLE I
Synthesis and Some Physical Properties of PAIs (6a–f)

5,5-Disubstituted hydantoin	Polymer	Yield (%)	η_{inh} (dL/g) ^a	$[\alpha]_{\text{D}}^{25}$ ^b
5a	6a	83	0.28	–6.0
5b	6b	91	0.31	–6.2
5c	6c	86	0.44	–5.9
5d	6d	87	0.32	–7.0
5e	6e	87	0.37	–5.1
5f	6f	94	0.30	–5.2

^{a,b} Measured at a concentration of 0.5 g /dL in DMF at 25°C.

TABLE II
Elemental Analysis of Aromatic PAIs (6a-f)

Polymer	Formula		C%	H%	N%
6a	C ₄₃ H ₂₈ N ₄ O ₈	calcd	70.87	3.87	7.68
	(728.27) _n	found	71.50	4.20	6.80
6b	C ₄₅ H ₃₂ N ₄ O ₈	calcd	71.42	4.26	7.40
	(756.76) _n	found	72.20	4.90	6.90
6c	C ₃₃ H ₂₄ N ₄ O ₈	calcd	65.56	4.00	9.26
	(604.57) _n	found	66.80	4.80	8.90
6d	C ₃₄ H ₂₆ N ₄ O ₈	calcd	66.01	4.23	9.05
	(618.59) _n	found	67.80	4.50	9.10
6e	C ₃₅ H ₂₈ N ₄ O ₈	calcd	66.45	4.46	8.85
	(732.62) _n	found	67.50	4.70	9.00
6f	C ₃₆ H ₂₈ N ₄ O ₈	calcd	67.07	4.37	8.69
	(644.63) _n	found	69.00	4.60	7.30

herent viscosities in a range from 0.28–0.44 dL/g. PAIs derived from monomer (4) may range in color from cream or off-white to brown. All of the resulting polymers show optical rotation and are optically active. The structures of these polymers were confirmed as PAIs by means of FTIR spectroscopy and elemental analyses (Table II).

The representative FTIR spectrum of PAIs 6a is shown in Figure 3. The polymers show absorption bands around 1780–1670 cm⁻¹ due to amide and imide carbonyl linkages. Absorption bands around 1390–1360 cm⁻¹ and 740–710 show the presence of the imide heterocycle in this polymer. The others spectra show a similar pattern. The elemental analysis values of the resulting polymers are in good agreement with the calculated values for the proposed structures (Table II).

Comparing inherent viscosities of this method with the previously method^{28,29} show that the internal heat generation of both the solvent and the monomers under the microwave irradiation was much more effective for the progress of polycondensation, producing PAIs having higher viscosity value in a shorter polymerization time, than the conventional external heating.

The solubility of PAIs 6(a-f) were investigated as 0.01 g of the polymeric sample in 2 mL of solvent. All of the polymers are soluble in organic solvents such as DMF, DMAc, and DMSO at room temperature, and

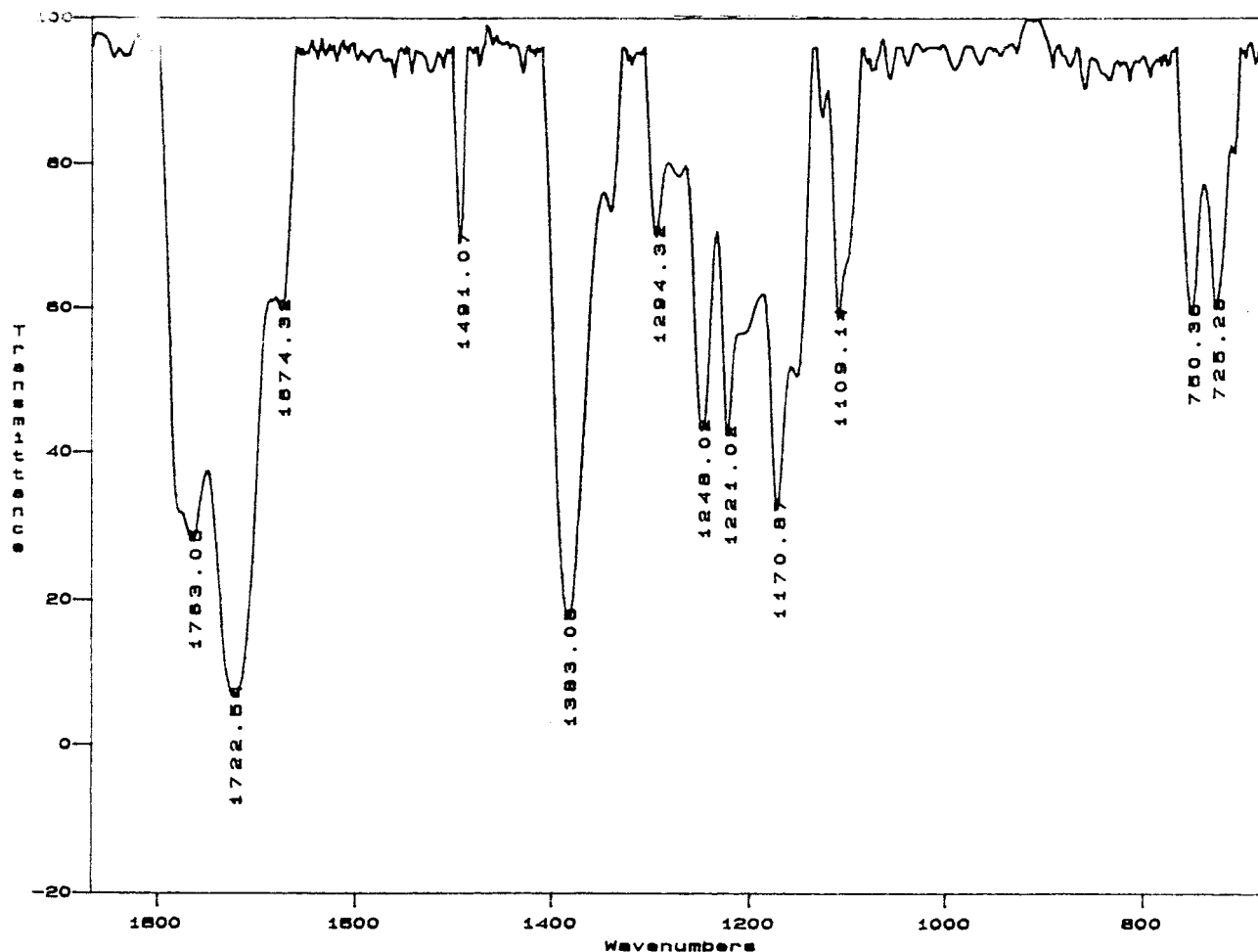


Figure 3 FTIR spectrum of PAIs (6a).

TABLE III
Thermal Behavior of Aromatic PAIs

Polymer	Decomposition Temperature(°C) T_5^a	Char yield(%) ^b
6a	255–260	14.0
6c	245–250	7.0

^a Temperature at which 5% weight loss was recorded by TGA at heating rate of 10°C/min in N₂

^b Percentage weight of material left undecomposed after TGA analysis at maximum temperature 600°C in N₂.

are insoluble in solvents such as chloroform, methylene chloride, methanol, ethanol, and water.

Thermal properties

The thermal properties of PAIs (**6a**, **6c**) were evaluated by means of TGA/DTG in nitrogen atmosphere. All of the polymers show similar decomposition behavior (Figs. 4–5). For **6a**, it started to decompose at 255°C, had lost 5% of its weight due to decomposition by 260°C, and by 600°C only had 14.0% of original weight. For **6c** it started to decompose at 245°C, had lost 5% of its weight due to decomposition by 250°C, and by 600°C only had 7% of original weight.

CONCLUSIONS

The present work has shown that *N,N'*-(Pyromellitoyl)-bis-*L*-phenylalanine diacid chloride (**4**) is an interesting monomer that contains both a pyrimillitimide rings as well as a chiral *L*-phenylalanine group. Thus, a series of new optically active PAIs (**6a–f**) were

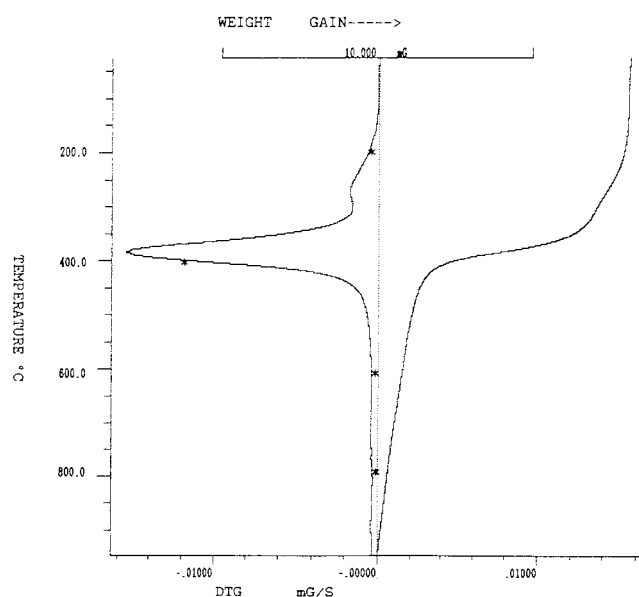


Figure 4 TGA and DTG thermogram of PAIs (**6a**).

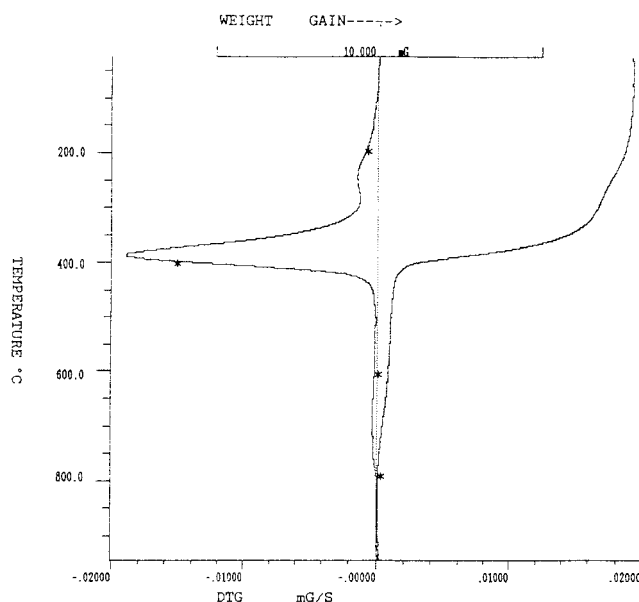


Figure 5 TGA and DTG thermogram of PAIs (**6c**).

able to be synthesized by microwave-assisted polycondensation reaction using the optically active monomer (**4**) with six different derivatives 5,5-disubstituted hydantoin (**5a–f**). These aromatic PAIs are optically active, are soluble in various organic solvents, and have good thermal stability. These resulting novel polymers have the potential to be used in a column chromatography technique for the separation of the enantiomeric mixtures. Furthermore, the above results demonstrate that microwave heating is an efficient method (shorter reaction time and high efficiency of energy) for the polycondensation reactions. We are currently using this method for the synthesis of novel polymers and modification of polymers.

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