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Synthesis of Novel Optically Active Poly(amide-imide)s with Benzophenone and *L*-Alanine Moieties

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ABSTRACT

3,3',4,4'-Benzophenonetetracarboxylic dianhydride (4,4'-carbonyldiphthalic anhydride) **1** was reacted with *L*-alanine **2** in a mixture of acetic acid and pyridine (3:2) at room temperature, then was refluxed at 90–100 °C and the resulting imide-acid **4** was obtained in quantitative yield. The compound **4** was converted to the diacid chloride **5** by reaction with thionyl chloride. The polymerization reaction of the imide-acid chloride **5** with benzidine **6a**, 4,4'-diaminodiphenyl methane **6b**, 1,5-diaminoanthraquinone **6c**, 2,6-diamino pyridine **6d** and 4,4'-sulfonyl dianiline **6e** was carried out in DMAc solution in the presence of pyridine. 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 IR spectroscopy, elemental analyses and specific rotation. Some structural characterization and physical properties of this optically active poly(amide-imide)s are reported.

Key Words: poly(amide-imide)s, *L*-alanine, optically active polymers, polycondensation reaction, benzophenone group

INTRODUCTION

Imide polymers and copolymers are a well-established class of polymers that show a good combination of mechanical, electrical, and thermal properties [1–2].

Since fully aromatic polyamides are intractable materials that do not melt before thermally decomposition and do not dissolve in any organic solvent, copolyimides were developed as a valuable alternative in the 1960s [3–4]. Many attempts have been made since then to modify the chemical structure of polyimides and copolyimides, always with the aim of

minimizing the shortcomings of aromatic polyimides while preserving their outstanding properties to the largest extent [5–6].

Many approaches have been investigated in attempting to improve the solubility of aromatic polyamides include the addition of pendant groups to the polymeric backbone [7–8] and the incorporation of bulky [9–10] or flexible [11–12] units within the parent chain. This paper describes the synthesis and properties of poly (amide-imide)s (PAIs) prepared from aromatic diamines and 4,4'-carbonyl-bis (phthaloyl-*L*-alanine) diacid chloride **5**, as a chiral

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monomer containing preformed imide groups.

Monomers bearing carbonyl groups attract considerable attention because of the possibility to use them in the preparation of polyimides with enhanced solubility, thermal stability and improved processibility. The use of monomers containing preformed imide rings is one method of circumventing partially cross-linked, because it avoids high temperature curing cycles and handling unstable intermediates such as polyamic acids [13–14]. Thus we designed the above monomer with a preformed imide ring as an "enlarged" monomer containing an additional carbonyl group and two chiral *L*-alanine groups, which also allows to achieve chiral PAIs with appropriate molecular weight by conventional means at low temperature.

The synthesis and application of optically active polymers are the newly considerable topics that have been paid more attention recently, because polymers with chiral structures are biologically very important. Most of the natural polymers are optically active and have special chemical activities such as catalytic properties that exist in genes, proteins and enzymes. Some other applications could be listed as: (1) constructing chiral media for asymmetric synthesis, (2) chiral stationary phases for resolution of enantiomers in chromatographic techniques, (3) chiral liquid crystals in ferroelectrics and nonlinear optical devices [15–18]. These applications have caused more considerations to improve different synthetic procedure of optically active polymers. Optically active homopolymers as well as copolymers have been prepared and reported in the literature [19–29].

In a previous paper [30] we synthesized a series of novel optically active poly(amide-imide)s containing one group of hexafluoropropylidene as well as two groups of chiral *L*-leucines. The resulting polymers are optically active and have an inherent viscosities in a range of 0.09–0.29 dLg⁻¹.

EXPERIMENTAL

Materials

All chemicals were purchased from Fluka Chemical

Co., Aldrich Chemical Co. and Riedel-deHaen AG. 3,3',4,4'-benzophenonetetracarboxylic dianhydride (4,4'-carbonyldipthalic anhydride) **1** was obtained from Fluka Chemical Co. as electronic grade material and was used as received. *N,N'*-dimethylacetamide (DMAc) was stirred over BaO for 24 h followed by vacuum distillation. Benzidine **6a** and 4,4'-diaminodiphenyl methane **6b** were purified by recrystallization from ethanol and distilled water, respectively. 2,6-Diaminopyridine **6d** was purified by sublimation. The other diamines were used as obtained without further purification.

Techniques

Proton nuclear magnetic resonance (¹H NMR, 90 MHz) spectra were recorded on a Varian EM-390 instrument. Multiplicities of proton resonances are designated as singlet (s), doublet (d), triplet (t), quartet (q), multiplet (m) and broad (br). Tetramethylsilane (TMS) was used as an internal reference.

IR Spectra were recorded on Shimadzu 435 IR spectrophotometer. Spectra of solids were carried out using KBr pellets. Vibrational transition frequencies are reported in wavenumber (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 Cannon Fensk Routine Viscometer. Specific rotations were measured by a Perkin Elmer-241 Polarimeter.

Thermal gravimetric analysis (TGA) data for polymers were taken on a Stanton-650 TGA under a N₂ atmosphere at a rate of 10 °C/min.

Monomer Synthesis

4,4'-Carbonyl-bis(phthaloyl-*L*-alanine) or (*N,N'*-carbonyldipthaloyl)bisalanine **4**

Into a 100 mL round-bottomed flask (4.00 g, 1.24 × 10⁻² mol) of 3,3',4,4'-benzophenonetetracarboxylic dianhydride **1**, (2.212 g, 2.48 × 10⁻² mol) of *L*-alanine **2**, 40 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 it was refluxed for 4 h. The solvent was removed under reduced pressure and the residue was dissolved in 100 mL of

cold water, then the solution was decanted and 5 mL of concentrated HCl was added. A white precipitate was formed, filtered off and dried, to give 5.30g (92.0%) of compound 4; mp 262–264 °C, $[\alpha]_D^{25} = -43.00$ (0.05 g in 10 mL DMF); IR (KBr): 3600–2800 (m, br), 1780–1760 (m), 1740–1680 (s), 1660–1640 (m), 1580–1520 (m, br), 1400–1360 (s), 1300–1280 (w), 1260–1220 (m, br), 1160–1140 (m), 1100–1080 (m), 1025–1015 (m), 980 (w), 920–900 (w), 880–850 (w, br), 760–700 (m, br), 640–600 (w, br) cm^{-1} , ^1H NMR (DMSO- d_6 , TMS): δ 1.5–1.6 (d, 6H); 4.8–5.2 (q, 2H); 8.2–8.5 (m, 6H).

The elemental analysis results obtained as follows:

$\text{C}_{23}\text{H}_{16}\text{N}_2\text{O}_9$	C (%)	H (%)	N (%)
Calculated	59.47	3.47	6.03
Found	58.70	3.70	5.70

4,4'-Carbonyl-bis(phthaloyl-L-alanine) diacid chloride (N,N'-carbonyldipthaloyl) bisalanine diacid chloride 5

Into a 25 mL round-bottomed flask were placed (2.00 g, 4.31×10^{-3} mol) of compound 4 and 10 mL (16.4g, 0.138 mol) 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 for over night 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 (94.5%) of pale yellow crystals; mp 156–158 °C $[\alpha]_D^{25} = -34.00$ (0.05 g in 10 mL DMF); IR (KBr): 3600–3200 (w, br), 3000–2900 (w), 1800–1760 (m), 1740–1700 (s), 1670–1650 (m), 1490–1460 (s), 1290–1270 (w), 1260–1240 (w), 1200–1180 (w), 1160–1140 (w), 960–920 (w, br), 900–870 (w), 740–700 (m) cm^{-1} , ^1H NMR (DMSO- d_6 , TMS): δ 1.6–1.7 (d, 6H); 4.9–5.2 (q, 2H), 8.2–8.5 (m, 6H).

The elemental analysis results obtained as follows:

$\text{C}_{23}\text{H}_{14}\text{N}_2\text{O}_7\text{Cl}_2$	C (%)	H (%)	N (%)
Calculated	55.11	2.82	5.59
Found	55.20	3.10	5.80

Polymer Synthesis

The poly(amide-imide) 7d was synthesized by four different methods:

Method A

Into a 25 mL round-bottomed flask was placed a stirring bar and (0.1 g, 1.996×10^{-4} mol) of diacid chloride 5, (0.0218 g, 1.996×10^{-4} mol) of 2,6-diaminopyridine 6d and 1 mL of CHCl_3 . The mixture was heated at 75 °C for 12 h. During this time the polymer was precipitated and 1 mL of DMAc was added and the reaction mixture was heated at 120 °C for 12 h. The resulting polymer was poured into 50 mL of methanol. The polymer was filtered off, and dried, to yield 0.100 g (93%) of brown solid.

Method B

Diacid chloride 5 (0.100 g, 1.996×10^{-4} mol), 2,6-diaminopyridine 6d (0.0218g, 1.996×10^{-4} mol), and 1 mL DMAc were placed in a 25 mL round bottom flask. The reaction mixture was stirred and heated at 120 °C for 24 h, then was poured into 50 mL of methanol. The resulting polymer was filtered off and was dried, to yield 0.100 g (93%) of brown solid.

Method C

Into a 25 mL round-bottom flask fitted with a stirring bar was added diacid chloride 5 (0.1 g, 1.996×10^{-4} mol) and 2,6-diaminopyridine 6d (0.0218 g, 1.996×10^{-4} mol). Then the reaction mixture was heated at 60 °C for 4 h and 120 °C for 0.5 h. Intensive stirring and dynamic vacuum enabled the removal of HCl. The solid was poured into 50 mL of methanol. The resulting polymer was filtered off, and dried to yield 0.08 g (74.6 %) brown solid.

Method D

Into a 25 mL round-bottomed flask fitted with a stirring bar was placed of 2,6-diaminopyridine 6d (0.0218 g, 1.996×10^{-4} mol) and 0.6 mL of DMAc. The mixture was cooled in an ice-water bath. To this solution (0.0316 g, 3.992×10^{-4} mol) of pyridine was added. Then (0.100 g, 1.996×10^{-4} mol) of solid diacid chloride 5 was added all at once. The polymerization proceeded as the acid chloride was dissolved. The

reaction mixture was stirred in ice-water bath for 1 h and the cooling bath was removed and the stirring was continued at room temperature for overnight and then it was heated at 80 °C for 12 h. The reaction mixture was poured into 50 mL of water. The precipitated polymer was collected by filtration and washed thoroughly with water and was dried at 80 °C for 8 h under vacuum to leave to 0.100g (93%) of brown solid.

Polymer 7a

IR (KBr): 3600–3200 (m, br), 3000–2800 (m, br), 1780–1760 (m), 1740–1700 (s), 1620–1580 (m), 1510–1480 (m), 1400–1360 (m), 1260–1240 (w), 1100–1080 (w), 1100–1080 (w), 1020–1000 (w), 820–800 (w), 730–700 (w) cm^{-1}

Polymer 7b

IR (KBr): 3600–3200 (m, br), 3000–2900 (w), 1780–1760 (w), 1740–1700 (s), 1630–1600 (w), 1520–1500 (m), 1390–1370 (m), 1300–1280 (w), 1260–1240 (w), 1110–1080 (w), 1020–1000 (w), 860–820 (w), 740–700 (w) cm^{-1} .

Polymer 7c

IR (KBr): 3600–3200 (m, br), 3000–2900 (w), 1780–1760 (w), 1740–1700 (s), 1640–1620 (w), 1600–1570 (m), 1520–1480(w), 1390–1370(w), 1260–1240 (m), 1180(w), 1080–1060 (w), 1040–1000 (w), 880 (w), 720–700 (w) cm^{-1} .

Polymer 7d

IR (KBr): 3600–3200 (m, br), 3000–2800 (w), 1780–1760 (w), 1740–1700(s), 1620–1570 (m, br), 1460–1440 (m), 1390–1370 (m), 1300–1280 (w), 1260–1240 (w), 1160–1140 (w), 1100–1080 (w), 1020–1000 (w), 1020–1000 (w), 730–710 (w) cm^{-1} .

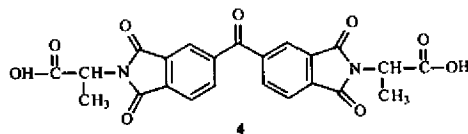
Polymer 7e

IR (KBr): 3600–3200 (m, br), 3000–2900 (w), 1780–1760 (w), 1740–1700 (s), 1620 (w), 1600–1580 (m), 1540–1520 (w), 1500–1480 (w), 1400–1360 (m), 1320–1280 (m), 1260–1240 (w), 1160–1140 (m), 1100–1080 (w), 1020–1000 (w), 840–820 (w), 740–680 (m, br) cm^{-1} .

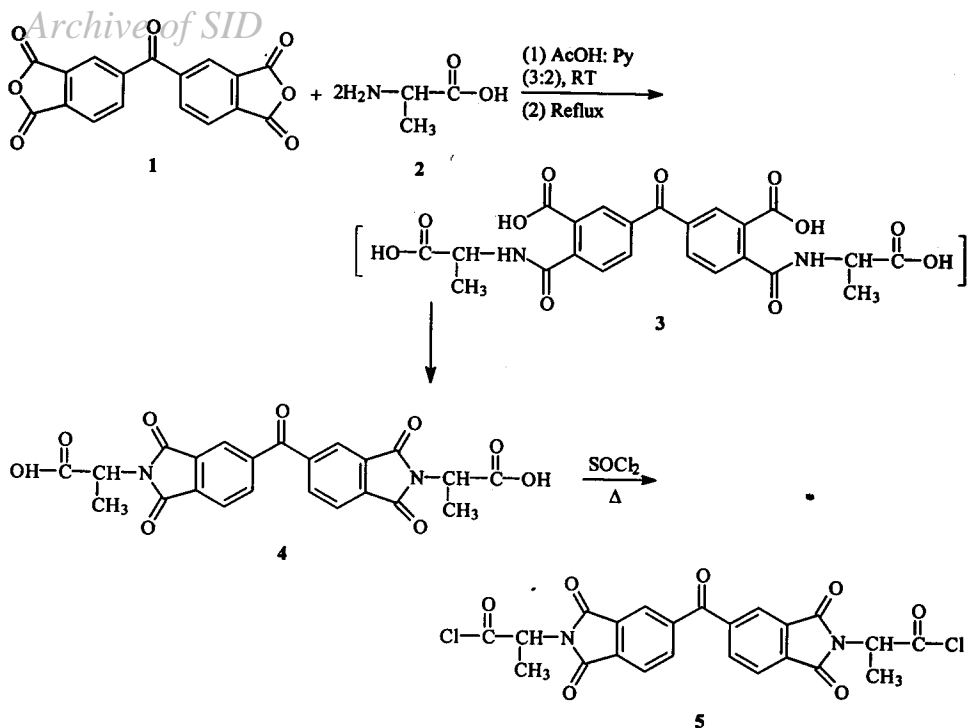
RESULTS AND DISCUSSION

Monomer Synthesis

4,4'-Carbonyl-bis(phthaloyl-*L*-alanine) diacid chloride **5** has been prepared in two-steps by refluxing of 4,4'-carbonyldiphthalic anhydride with two moles of *L*-alanine in acetic acid, following by reaction of the resulting diacids **4** with thionyl chloride, in literature [31–33]. We have prepared 4,4'-carbonyl-bis(phthaloyl-*L*-alanine) diacid chloride **5** in two-steps with a modified procedure as shown in Scheme 1. The asymmetric diacid **4** was synthesized by the condensation reaction of two equimolar of *L*-alanine 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 pale yellow solid in quantitative yield. The resulting asymmetric diacid **4** was converted to its diacid chloride derivative **5** by the reaction with thionyl chloride. The monomer **5** was purified by washing with *n*-hexane. The chemical structure and purity of the optically active monomers **4** and **5** were proved using elemental analysis, IR and ¹H NMR spectroscopic techniques. The ¹H NMR spectrum of compound **4** showed peaks at 1.5–1.6 ppm as a doublet, which was assigned to two CH₃ groups. Peaks between 4.8–5.2 ppm as a quartet was assigned to the CH(c) protons, which is a chiral centre. The peak between 8.2–8.5 ppm were assigned to aromatic protons and peak at 9.55 ppm was assigned to COOH proton.



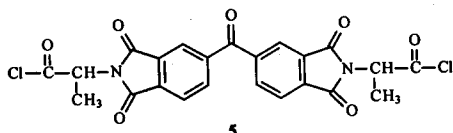
The IR spectrum of compound **4** showed a broad and strong peak between 2800–3600 cm^{-1} which was assigned to the COOH groups and two absorption bands at 1780 and 1700 cm^{-1} which are characteristic peaks for imide rings. Disappearance of strong acidic hydroxyl peak in IR spectrum of compound **5** confirmed a complete conversion of



Scheme 1

diacid **4** to diacid chloride **5**. On the other hand, because of the electron withdrawing character of the Cl group, the two carbonyl peaks of diacid chloride in comparison with its starting diacid, were shifted to higher frequency.

The ^1H NMR spectrum of compound **5** showed peaks as a doublet between 1.6–1.7 ppm for two CH_3 protons, a quartet peak at 4.9–5.2 ppm for two $\text{CH}(\text{c})$ protons. The peaks between 8.25–8.55 ppm were assigned to aromatic protons.



Polymer Synthesis

Poly(amide-imide)s **7a–7e** were synthesized by solution polycondensation reactions of an equimolar mixture of monomer **5** with five different diamines **6a–6e** in DMAc as shown in Scheme II. Poly(amide-imide)s **7d** were prepared from the monomer **5** and diamine **6d** by four different methods.

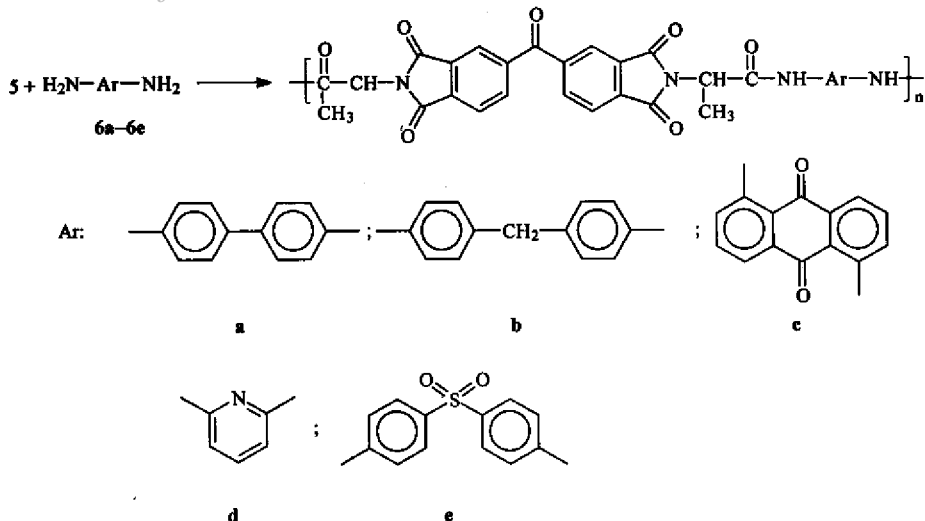
In method A, the polymerization was carried out in $\text{CHCl}_3/\text{DMAc}$ solution at high temperature, without any scavenger such as pyridine, and the resulting polymer has low inherent viscosity.

In method B, the polymerization was performed in DMAc solution at high temperature, in the absence of acid scavenger, and the resulting polymer has still low inherent viscosity.

In method C, the polymerization was carried

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Scheme II

out under bulk condition at higher temperature, and in vacuum, and the polymerization was carried out in DMAc solution at low temperature and then was heated at higher temperature in presence of pyridine, and the resulting polymer has higher inherent viscosity (Table 1). According to these results we selected the method D for the synthesis of the other poly(amide-imide)s.

Table 1. Synthesis and some physical properties of poly(amide-imide) 7d.

Diamine	Method	Yield (%)	η_{inh} (dL/g) ^a	Remarks ^b
6d	A	93	0.153	P
6d	B	93	0.115	S
6d	C	75	0.114	-
6d	D	93	0.330	P

(a) Measured at a concentration of 0.5 g/dL in DMF at 25 °C; (b) Appearance of the polymerization mixture: S, homogeneous solution throughout the reaction; and P, precipitation during the reaction.

Polymer Characterization

Poly(amide-imide)s derived from monomer 5 may range in colour from cream or off-white to pale-yellow, except polymers 7c and 7d which have an

Table 2. Synthesis and some physical properties of aromatic poly(amide-imide)s 7a-7e.

Diamine	Polymer	Yield (%)	η_{inh} (dL/g) ^a	$[\alpha]_D^{25}$ ^b	Remarks ^c
6a	7a	92	0.200	-33.75	P/S
6b	7b	90	0.200	-12.75	P/S
6c	7c	95	0.320	-	P/S
6d	7d	93	0.330	-	P/S
6e	7e	85	0.180	-35.00	P/S

(a) Measured at a concentration of 0.5 g/dL in DMF at 25 °C; (b) Measured at a concentration of 0.5 g/dL in DMF at 25 °C; (c) Appearance of the polymerization mixture: S, homogeneous solution throughout the reaction; and P, precipitation during the reaction; P/S, precipitation at the first step and homogeneous solution of the second one.

Table 3. Elemental analysis of poly(amide-imide)s 3a–3e.

Polymer	Formula		Elemental analysis (%)			Moisture intake (%) ^a
			C	H	N	
7a	(C ₃₅ H ₂₄ N ₄ O ₇) _n (612) _n	Calcd	68.01	3.95	9.15	2.50
		Found	66.30	4.50	8.50	
		Corr ^b	67.96	4.38	8.71	
7b	(C ₃₆ H ₂₆ N ₄ O ₇) _n (626) _n	Calcd	68.09	4.18	8.95	1.50
		Found	67.00	5.10	7.80	
		Corr ^b	68.00	5.02	7.92	
7c	(C ₃₇ H ₂₂ N ₄ O ₉) _n (666) _n	Calcd	66.65	3.33	8.41	1.60
		Found	65.50	3.90	8.80	
		Corr ^b	66.55	3.83	8.94	
7d	(C ₂₉ H ₁₉ N ₅ O ₇) _n (537) _n	Calcd	62.07	3.56	13.03	3.20
		Found	60.00	4.00	11.10	
		Corr ^b	61.92	3.87	11.45	
7e	(C ₃₅ H ₂₄ N ₄ O ₅ S) _n (676) _n	Calcd	62.13	3.58	8.25	2.80
		Found	60.40	4.10	7.50	
		Corr ^b	61.97	3.99	7.70	

(a) Moisture intake (%) = $(W_1 - W_2) \times 100 / W_1$. W_1 = weight of polymer sample after standing at room temperature and W_2 = weight of polymer sample after dried in vacuum at 100 °C for 10 h; (b) For C and N: corrected value = found value \times (100% + moisture intake %). For H: corrected value = found value \times (100% - moisture intake %).

intense orange colour and pale orange colour, respectively. According to this deep colour, the polarized light could not transmit through the polymer solution, and therefore their optical rotations were undefined (Table 2). The other new polymers show rotation, and therefore they are optically active.

The structures of these polymers were confirmed as poly(amide-imide)s by means of IR spectroscopy and elemental analysis. The IR spectra of all polymers showed absorptions around 3300 cm^{-1} (N–H) and two overlapped carbonyl (amide and imide C=O) absorptions at 1720 and 1780 cm^{-1} . All of them exhibited strong absorbance at 1370–1380 cm^{-1} and 710–720 cm^{-1} , which show the presence of the imide heterocycle in these polymers. The polymer 7e showed characteristic absorptions at 1300 and 1120 cm^{-1} due to the sulphone moiety (SO₂ stretching). The elemental analysis values of the resulting polymers are in good agreement with the calculated values for the proposed structures of the resulting polymers. Since

poly (amide-imide)s tend to absorb moisture, we have corrected this factor for the elemental analyses data (Table 3).

The solubilities of poly (amide-imide)s are listed in Table 4. Most of the polymers are soluble in organic solvents such as DMF, DMAc and DMSO at room temperature, and they are insoluble in solvents such as chloroform, methylene chloride, methanol, ethanol and water.

Thermal Properties

The thermal decomposition temperatures were evaluated by means of DSC and TGA, respectively. Table 5 summarizes the thermal properties of all the poly(amide-imide)s.

The poly(amide-imide)s exhibited good resistance to thermal decomposition up to 227–250 °C in nitrogen and began to decompose gradually above that temperature. The temperature of 5% weight loss for all the polymers ranged from 227 to 251 °C and the

Table 4. Solubility of poly(amide-imide)s 7a–7e.

Solvents	7a	7b	7c	7d	7e
DMAc	+	+	+	+	+
DMF	+	+	+	+	+
THE	–	–	–	–	–
DMSO	+	+	+	+	+
MeOH	–	–	–	–	–
EtOH	–	–	–	–	–
CHCl ₃	–	–	–	–	–
CH ₂ Cl ₂	–	–	–	–	–
H ₂ O	–	–	–	–	–

+: Soluble at room temperature, –: Insoluble.

residual weight for these polymers at 600 °C ranged from 49.92 to 59.47 % in nitrogen.

CONCLUSION

The present work has shown that 4,4'-carbonyl-bis (phthaloyl-*L*-alanine) diacid chloride **5** is an interesting monomer, which contains both benzophenone-imide group as well as chiral *L*-alanine groups. Thus, a series of new optically active poly(amide-imide)s having inherent viscosities of 0.15–0.33 dL/g were synthesized by the solution polycondensation reaction of the novel optically active monomer, 4,4'-carbonyl-bis (phthaloyl-*L*-alanine) diacid chloride **5** with some aromatic diamines. These aromatic poly(amide-imides) are optically active and are readily soluble in

Table 5. Thermal behaviour of aromatic poly(amide-imide)s.

Polymer	Decomposition temperature °C T_5^a	Char yield (%) ^b
7a	237	53.81
7b	242	55.41
7c	251	59.47
7d	227	49.92

(a) Temperature at which 5% weight loss was recorded by TG at heating rate of 10°C/min in N₂. (b) Percentage weight of material left undecomposed after TG analysis at maximum temperature 600 °C in N₂.

various organic solvents and have good thermal stability. These polymers have potential to be used as packing materials in column chromatography for the separation of enantiomers. Although the inherent viscosities are not very high, but attempts for a modification in order to synthesize polymers with higher inherent viscosities using other polymerization techniques are under investigation.

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