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International Journal of Polymeric Materials

Publication details, including instructions for authors and subscription information:

<http://www.informaworld.com/smpp/title~content=t713647664>

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Shantilal B. Oswal^a; Naved I. Malek^b; Ashesh K. Pandya^c

^a Biochemistry Division, R&D Span Diagnostic Limited, Udhna, Surat, India ^b ASHD, S. V. National Institute of Technology, Surat, India ^c R&D Division, Calyx Chemicals & Pharmaceuticals, Dombivli East, Dist-Thane, India

To cite this Article Oswal, Shantilal B. , Malek, Naved I. and Pandya, Ashesh K.(2009) 'Interfacial Polymerization of Linear Aromatic Poly(ester amide)s', International Journal of Polymeric Materials, 58: 4, 202 – 216

To link to this Article: DOI: 10.1080/00914030802639973

URL: <http://dx.doi.org/10.1080/00914030802639973>

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Interfacial Polymerization of Linear Aromatic Poly(ester amide)s

Shantilal B. Oswal,¹ Naved I. Malek,² and
Ashesh K. Pandya³

¹Biochemistry Division, R&D Span Diagnostic Limited, Udhna,
Surat, India

²ASHD, S. V. National Institute of Technology, Surat, India

³R&D Division, Calyx Chemicals & Pharmaceuticals, Dombivli East,
Dist-Thane, India

Linear aromatic poly(ester amide)s (PEAs) have been synthesized by interfacial polycondensation (IPC) of aromatic diamidoacid chloride: 2-([4-([2-(chlorocarbonyl)phenyl]amino) carbonyl] benzoyl]amino) benzoyl chloride (2CCBC), with ethylene glycol, bisphenol A, resorcinol, 4,4'-bis(4-hydroxybenzilidene)diaminobenzanilide and 4,4'-bis(4-hydroxy benzilidene)-m-phenylenediamine in chloroform/water system employing phase-transfer-catalyst. The aromatic diamidoacid chloride has been prepared by condensation of terephthaloyl chloride with anthranilic acid. These polymers were characterized by elemental analysis, FTIR, ¹H-NMR, solubility studies, intrinsic viscosity and TGA analysis. The polyester-amides so obtained show good thermal stability.

Keywords: interfacial polycondensation, intrinsic viscosity, phase transfer catalysis, poly(ester amide)s, TGA,

INTRODUCTION

The development of processable high-performance polymers with increased thermal stability, heat resistance and good mechanical properties have become an important problem [1]. In an attempt to prepare polymers of high glass transition temperature T_g, Imai et al. [2,3] synthesized poly(ester amide)s (PEAs) of high molecular weight and

Received 30 October 2008; in final form 8 November 2008.

Address correspondence to Shantilal B. Oswal, Biochemistry Division, Span Education Research Foundation, R&D Span Diagnostic Limited, 173-B, New Industrial Estate, Udhna, Surat – 394210, Gujarat, India. E-mail: oswalsl@yahoo.co.uk

investigated their properties in detail. PEAs have been the object of extensive investigation during the last few decades.

The PEAs have special structure due to the regularity of ester and amide groups in the same polymer chain, which gave them the properties intermediate between the polyester and the polyamide [4]. The rigidity due to the double-bond character of the amide group coupled with extensive hydrogen bonding influences the ordering of PEAs [5]. Apart from high thermal stability, many PEAs also exhibited liquid crystalline properties [6–10], enhanced by intermolecular hydrogen bonding between amide–amide and amide–ester moieties. Hydrogen bonds in polymers have a real influence on their physical and chemical properties, such as melting temperature, reactivity, solubility, adhesion or thermal stability. They also have a strong influence on the mechanical properties of the polymers. In the PEAs, there are two possibilities for hydrogen-bond formation, i.e., the amide-amide bond, as observed in polyamides, and also the amide-ester hydrogen bond, namely the bond between the hydrogen of the amide group and the carbonyl oxygen of the ester group [11–13]. These two possibilities are determined by the position of the ester and the amide groups along the polymer chains [14]. These materials also have technical potential because of high modulus fiber-forming properties, and as engineering polymers [15,16].

Further, PEAs constitute a new series of thermoplastic polymers that can combine a high technical performance with a good biodegradability. Bayer has recently produced the BAK 1095 and BAK 2195 polymers, which appear to have a wide range of applications as biodegradable materials [17]. Thus, PEAs have emerged as promising biodegradable materials with good mechanical and thermal properties, as well as facile processing and degradation susceptibility [18]. Considering all these important aspects, it was decided to undertake systematic studies to synthesize new PEAs and to investigate their properties in detail [19]. In this paper we report synthesis of new PEAs by interfacial polycondensation and their characterization through elemental analysis, solubility studies, intrinsic viscosity measurements, FTIR, $^1\text{H-NMR}$, and TGA analysis.

EXPERIMENTAL

Materials

4,4'-Diamino benzanilide from ATUL, Valsad, and terephthalic acid (TPA), p-amino benzoic acid, p-hydroxybenzaldehyde, m-phenylenediamine, bisphenol A (BPA), resorcinol (RES), ethylene glycol (EG), citramide (cetyltriethylammonium bromide), thionyl chloride, pyridine, and chloroform, all from SRL, Mumbai, were used as received.

Terephthaloyl chloride (TPCl) was prepared by the reaction of TPA with excess thionyl chloride, in the presence of a few drops of pyridine as catalyst. It was purified by crystallization from n-hexane.

Measurement

Elemental analysis (C, H, and N) for all PEAs was performed using a Carlo Erba Elemental Analyzer. Solubility of PEAs was determined for powdered samples in excess of solvent. Viscosity measurements of the polymer solutions were carried out with an Ubbelohde viscometer at 30°C using a thermostated waterbath. FTIR spectra of the monomers and all polymers have been scanned in KBr pellets on a Perkin Elmer IR Spectrometer. The ¹H-NMR spectra were taken in DMSO-d₆ as a solvent on Bruker DPX-200 Spectrometer at 200 MHz (RSIC, CDRI, Lucknow) with a sweep time of 10 min at room temperature. The internal reference used was TMS. Thermogravimetric analysis was carried out with a Mettler TA 4000 system (Switzerland). All runs were carried out under a flow of dry N₂ atmosphere at a heating rate of 10°C/min in the temperature range from 50°C to 700°C.

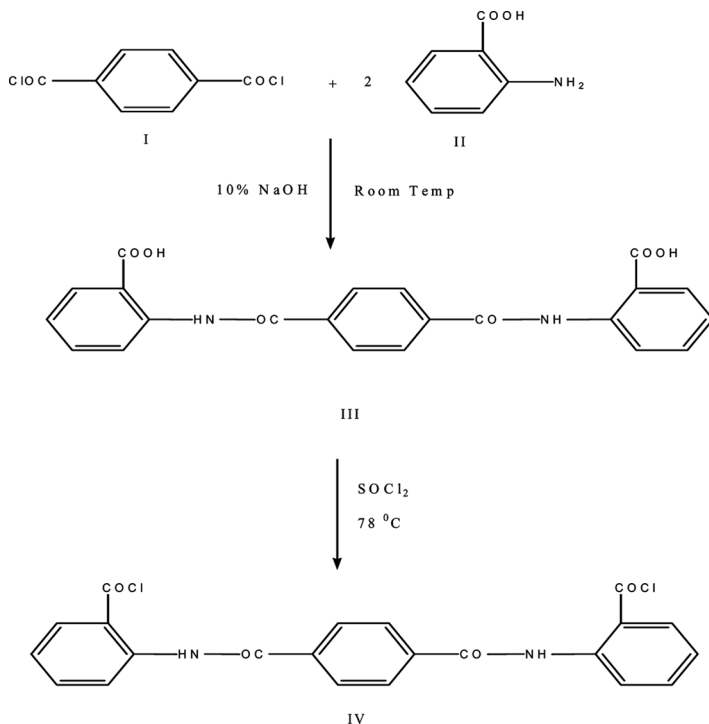
Monomer Synthesis

2-[[4-[[2-(chlorocarbonyl)phenyl]amino]carbonyl]benzoyl]amino]benzoyl-chloride (2CCBC)

It was synthesized in two steps as shown in Scheme 1.

2-[[4-[[2-(carboxyphenyl)amino]carbonyl]benzoyl]amino]benzoic acid (2CPBA, III)

It was synthesized from terephthaloyl chloride (I) and anthranilic acid (II) [20]. Anthranilic acid (0.02 mol) was dissolved in 50 ml of 10% NaOH solution. Terephthaloyl chloride (0.01 mol) was added in five portions to the solution. The solution was transferred to a beaker after adding a few grams of crushed ice to the solution. Concentrated HCl was slowly added with stirring until the mixture was acidic to congo-red paper. To remove a small contamination of TPA from the precipitate, it was washed with cold water and placed in a small beaker covered with watch-glass containing 20 ml of CCl₄. It was boiled gently for 10 min. The product was filtered by gentle suction. The product 2CPBA (III) was recrystallized from boiling water and dried in a vacuum desiccator. FTIR (in cm⁻¹): 3200-2500 (O-H stretch, carboxylic acid), 1680 (>C=O stretch), 3060 (C-H stretch, aromatic), 1610, 1510 and 1420 (C=C stretch, aromatic), 750 (C-H bend, 1,2-substituted benzene), 1590 (N-H bend), 3480 and 3380 (N-H stretch). ¹H-NMR (Figure 1, DMSO-d₆, 200 MHz, TMS,



SCHEME 1 Synthesis of 2-[[4-({2-(chlorocarbonyl)phenyl}amino)carbonyl]benzoyl]amino}benzoyl-chloride (2CCBC).

δ ppm): 12.25 (s, 2H, -COOH), 8.31 (s, 2H, -CO-NH), 8.02–8.28 (m, 4Ar-H, terephthaloyl moiety), 7.21–7.73 (m, 8Ar-H, anthranilic acid moiety).

2-[[4-({2-(chlorocarbonyl)phenyl}amino)carbonyl]benzoyl]amino}benzoyl Chloride (2CCBC) (IV)

2CPBA (III) (0.01 mol) was placed in a dry round-bottom flask. Thionyl chloride (0.2 mol) was added through the top of the condenser. The reaction mixture was refluxed at 78 °C for 2 h. The excess of thionyl chloride was distilled off and the dry product 2CCBC (IV) was recovered from the flask. The yield was about 85% and it was recrystallized from DMF.

4,4'-Bis(4-hydroxy benzylidene)Diaminobenzanilide (BHDABA)

It was synthesized from reaction of p-hydroxy benzaldehyde (V) and 4,4'-bis(4-hydroxy benzylidene)diamino benzanilide (DABA, VI)

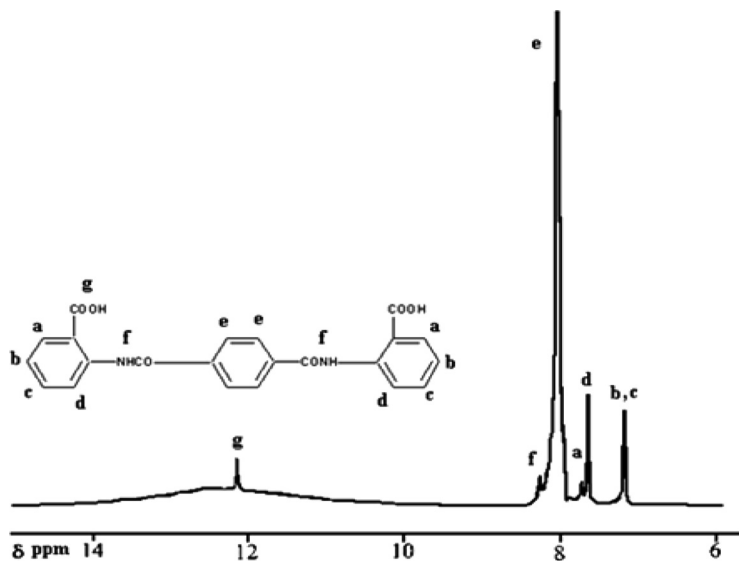
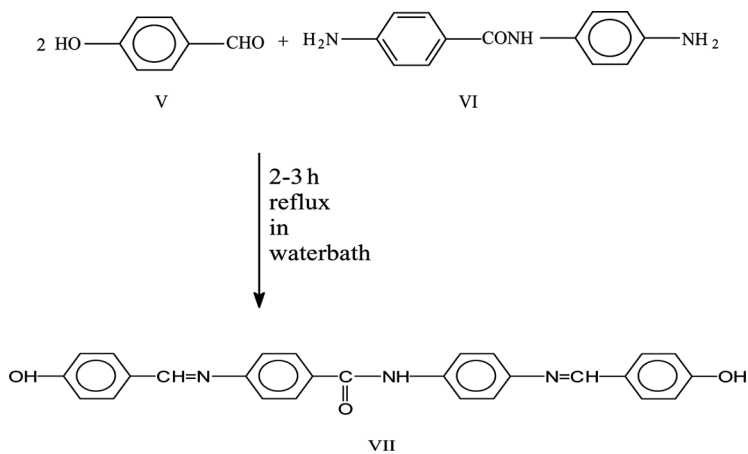


FIGURE 1 $^1\text{H-NMR}$ spectrum of 2CPBA in DMSO-d_6 at 200 MHz.

(Scheme 2). Ethanolic solution of p-hydroxy benzaldehyde (0.02 mol) was mixed with ethanolic solution of DABA (0.01 mol) and refluxed over a water bath for 2–3 h. The reaction mixture was poured into crushed ice. The yellow precipitate was filtered, washed with



SCHEME 2 Synthesis of 4,4'-bis(4-hydroxy benzylidene)diamino benzanilide (BHBDA).

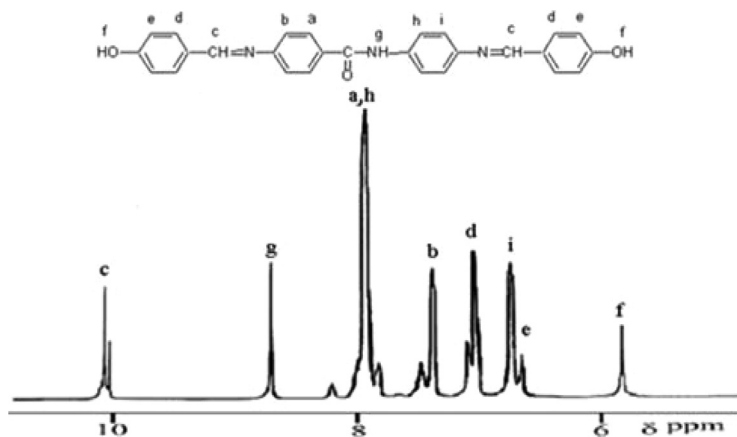


FIGURE 2 $^1\text{H-NMR}$ spectrum of BHBDA in DMSO-d_6 at 200 MHz.

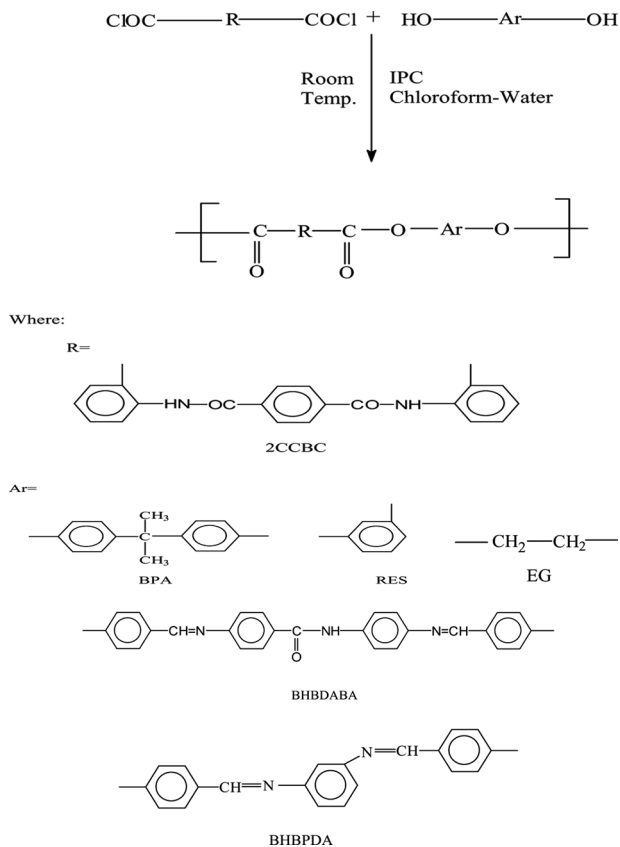
methanol and distilled water and then dried. The product BHBDA (VII) was purified by reprecipitation from chloroform-methanol (yield 84%, mp 251°C). FTIR (in cm^{-1}) 3400-3200 (broad O-H stretch), 1605 ($-\text{CH}=\text{N}$), 1640 ($> \text{C}=\text{O}$ stretch), 1530 (N-H bend), 3330 and 3290 (N-H stretch), 1590, 1510 and 1450 ($\text{C}=\text{C}$ stretch, aromatic). $^1\text{H-NMR}$ (Figure 2, DMSO-d_6 , 200 MHz, TMS, δ ppm) 5.63 (s, 2H, -OH), 10.25 (s, 2H, $-\text{CH}=\text{N}$), 8.48 (s, H, $-\text{CO-NH}$), 6.50–8.01 (m, 16 Ar-H).

4,4'-Bis(4-hydroxy benzilidene)-m-phenylenediamine (BHPDA)

It was synthesized by reacting p-hydroxy benzaldehyde (V) with m-phenylenediamine (VIII) (Scheme 3). Ethanolic solution of p-hydroxy benzaldehyde (0.02 mol) and ethanolic solution of m-phenylenediamine (0.01 mol) were mixed and refluxed over a water bath for 2–3 h. The reaction mixture was poured into crushed ice. The yellowish-green precipitate was filtered, washed with methanol and distilled water and then dried. The product BHPDA (IX) was purified by re-precipitation from chloroform-methanol (yield 90%, mp 164°C). FTIR (in cm^{-1}): 3390-3200 (broad O-H stretch), 1600 ($-\text{CH}=\text{N}$), 1630, 1510 and 1445 ($\text{C}=\text{C}$ aromatic), 920, 750 and 720 (C-H bend, 1,3-substituted benzene), 835 (C-H bend, 1,4-substituted benzene). $^1\text{H-NMR}$ (Figure 3, DMSO-d_6 , 200 MHz, TMS, δ ppm): 5.85 (s, 2H, -OH), 9.87 (s, 2H, $-\text{CH}=\text{N}$), 6.74–8.07(m, 12Ar-H).

Polymer Synthesis

PEAs were synthesized by interfacial polycondensation of 2CCBC with each of the different diols in a chloroform-water interface system and



SCHEME 4 Synthesis of poly(ester amide)s.

dissolved in 10 ml of water were added. The organic phase was prepared by dissolving 2CCBC (0.0125 mol) in 35 ml of chloroform. The speed of the stirrer was raised to the maximum value, and the organic phase was introduced rapidly. Mixing was continued thereafter for 5 min. At the end of 5 min period stirring (which was still in progress) an excess of acetone was added to the beaker. After one minute of gentle stirring, the polymer was filtered off and washed with acetone to remove unreacted monomers, chloroform, excess alkali and salts. The polymerized product was dried to constant weight in a vacuum oven at 40°C. Other PEAs were synthesized from 2CCBC and the diols RES, EG, BHDABA, and BHPDA adopting the same procedure.

RESULTS AND DISCUSSION

Conditions for Poly(ester amide) Synthesis

In the present investigation interfacial polycondensation of diamidoacid chloride with aromatic or aliphatic diol have been used. It has been observed [21–23] that the polymerization reaction is influenced by solvent system, catalytic and surfactant additives. The choice of the organic solvent is more important since it will affect several other polymerization factors such as the potential partition of reactants between the two phases, the diffusion of the reactants, reaction rate and the solubility, swelling or permeability of the growing polymer [21]. It has been reported [24] that attempts to prepare polyester amides with high molecular weights by interfacial polymerization in a nonpolar solvent-water system failed because of early polymer precipitation. Therefore, inert polar solvents that are water-immiscible or at least partially water-soluble were chosen for interfacial polycondensation, as shown in Table 1, for the synthesis of representative polyester-amide (2CCBC-BPA) using identical reaction conditions. It can be seen from Table 1 that the polyester amides with chloroform-water system have highest inherent viscosity η_{inh} and yield; therefore we have selected chloroform-water system for the present work.

Effect of Catalytic and Surfactant Additives

Surface-active agents are known to enhance the degree of polymerization in the interfacial production of polymers. This enhancement is

TABLE 1 Effect of Different Interphase Systems**

Interphase system	Phase-transfer catalyst	Yield %	η_{inh} (dl/g)*
Chloroform-water	Citramide	90	0.187
Toluene-water	Citramide	16	0.124
Benzene-water	Citramide	17	0.113
Cyclohexane-water	Citramide	68	0.130
Dichloroethane-water	Citramide	45	0.141
Nitrobenzene-water	Citramide	73	0.144
Chloroform-water	without catalyst	55	0.133
Chloroform-water	SDS	73	0.175
Chloroform-water	Tween 20	86	0.181

*in DMF at 30°C.

**Phase volume ratio: 35 ml organic solvent/85 ml H₂O, Monomers: 2CCBC and BPA each 0.0125 mol, NaOH: 0.025 mol, Phase transfer catalyst: 0.25 mg, Temp: 30°C, Time: 5 min, Stirring: continuous and vigorous.

brought about by improved mixing, increased interfacial area and consequently increased contact of the reactants. Sometimes IPC as type of nucleophilic displacement reaction is carried out with phase-transfer catalyst [25], generally small symmetric quaternary ammonium cations [22,23]. It has been suggested that both surfactant and catalytic functions could be combined through the use of cationic surfactants (as quaternary ammonium compounds) that could also be used in the transfer of monomer between phases.

We have employed three surfactants: (i) citramide as a cationic surfactant, (ii) sodium dodecyl sulphate (SDS) as an anionic surfactant, and (iii) Tween 20 as a nonionic surfactant, for the synthesis of polyester amide from 2CCBC and BPA using chloroform/water interface system as listed in Table 1. The results showed that the use of citramide gave the highest inherent viscosity and yield. The inherent viscosity of polymers is in the sequence: Citramide > Tween 20 > SDS > without catalyst. Therefore, in the present work, citramide was used as a cationic surfactant and phase-transfer catalyst for the synthesis of all the polyester amides.

Yield and General Appearance

Yields of the different polymer samples were in the range from 67 to 93%. All the poly(ester amide)s are solid powders and colored compounds depending upon the types of diol involved.

Elemental Analysis and Solubility Behavior

The results of elemental analysis of our poly(ester amide)s are given in Table 2. The observed percentages of C, H, and N in present poly(ester amide)s are in fairly good agreement with those theoretically calculated based on the repeat units in the chain.

Solubility test in different solvents reveals that the poly(ester amide)s P2C1 and P2C4 are soluble in DMF, THF, and dioxane while P2C3 is soluble in DMSO and chloroform. All the polymers are totally insoluble in carbon tetrachloride, methanol, ethanol, acetone, benzene, toluene, isopropanol and water.

Viscosity Measurements

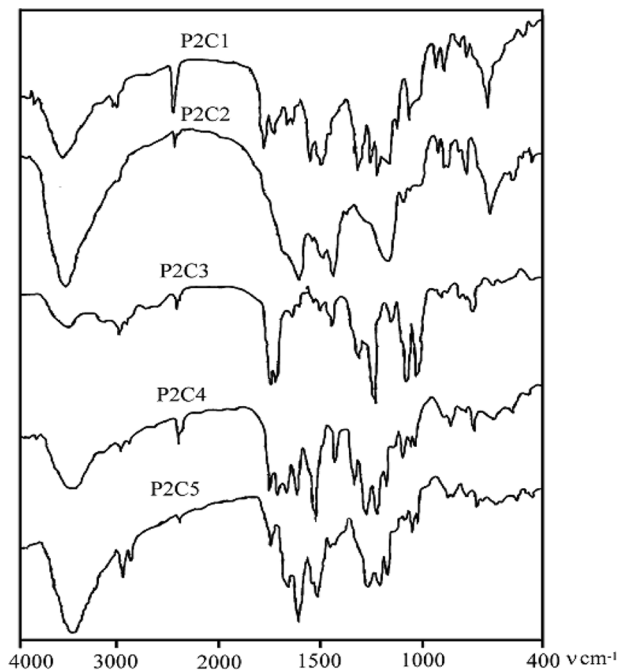
Viscosities of P2C1, P2C3 and P2C4 have been determined in DMF at 30°C. The average values of intrinsic viscosity for P2C1, P2C3 and P2C4 are 0.187, 0.122 and 0.194 dl g⁻¹ respectively.

TABLE 2 Elemental Analysis of the Poly(ester amide)s

Polymer		Elemental analysis		
		C (%)	H (%)	N (%)
P2C1 (2CCBC + BPA)	Calcd.	74.50	4.70	4.70
	Found	74.32	4.45	4.59
P2C2 (2CCBC + RES)	Calcd.	70.29	3.77	5.86
	Found	70.11	3.72	5.73
P2C3 (2CCBC + EG)	Calcd.	66.98	4.19	6.51
	Found	66.87	4.01	6.36
P2C4 (2CCBC + BHBDABA)	Calcd.	73.23	4.11	8.72
	Found	72.96	3.95	8.61
P2C5 (2CCBC + BHBPDA)	Calcd.	73.68	4.09	8.19
	Found	73.44	4.02	8.08

Spectral Studies

FTIR of present poly(ester amide)s are shown in Figure 4. The formation of poly(ester amide) is confirmed from the typical characteristic

**FIGURE 4** FTIR spectrum of P2C1, P2C2, P2C3, P2C4 and P2C5.

bands observed at $1726 \pm 16 \text{ cm}^{-1}$ ($>\text{C}=\text{O}$ stretch in ester) and 1009 ± 7 , 1035 ± 32 and $1246 \pm 43 \text{ cm}^{-1}$ (C-O-C symmetric and asymmetric stretch), and $1650 \pm 40 \text{ cm}^{-1}$ ($>\text{C}=\text{O}$ stretch in amide) [26–28]. But, in P2C2, due to the overlap of $>\text{C}=\text{O}$ stretch of ester and amide, the absorption peak is observed as a broad peak centered around $1640 \pm 20 \text{ cm}^{-1}$.

The different types of poly(ester amide)s can be distinguished from each other by a few other characteristic absorption frequencies of the involved aliphatic and aromatic diols. The involvement of BPA moiety in P2C1 is indicated by a band at $1384 \pm 20 \text{ cm}^{-1}$ (isopropyl group [4,29]). The presence of resorcinol moiety in P2C2 is confirmed due to bands at 900 ± 20 , 760 ± 20 , 683 cm^{-1} (1,3-disubstituted benzene [4]). In poly(ester amide)s P2C3 the absorption band at $1473 \pm 13 \text{ cm}^{-1}$ indicates this incorporation of $-(\text{CH}_2)_n-$ of ethylene glycol moiety [4]. It is difficult to separately identify BHDABA and BHPDA as these two diols contain the same aromatic or $-\text{CH}=\text{N}$ - groups which are also present in the diamidoacid chloride (2CCBC) monomer. Further absorption frequency of the $-\text{CH}=\text{N}$ - group also overlaps with aromatic $(-\text{C}=\text{C}-)$ stretch frequency. The characteristic band for $-\text{CH}=\text{N}$ - in benzilidine group is generally observed at $1610\text{--}1580 \text{ cm}^{-1}$ [4,30].

$^1\text{H-NMR}$ of P2C1 and P2C4 are given in Figures 5 and 6. P2C1: A sharp singlet at $\delta 1.59 \text{ ppm}$ is indicative of 6H (h) of isopropyl unit [31,32]. The polymer repeat unit has overall 20 Ar-H, which appear

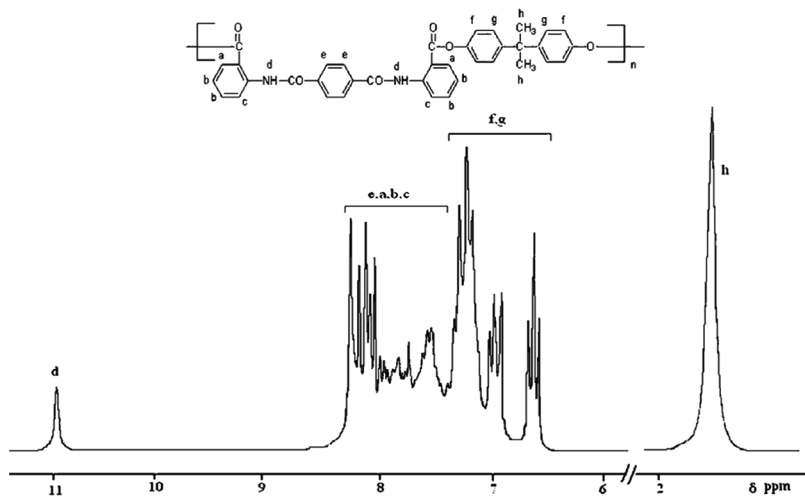


FIGURE 5 $^1\text{H-NMR}$ spectrum of P2C1 in DMSO-d_6 at 200 MHz.

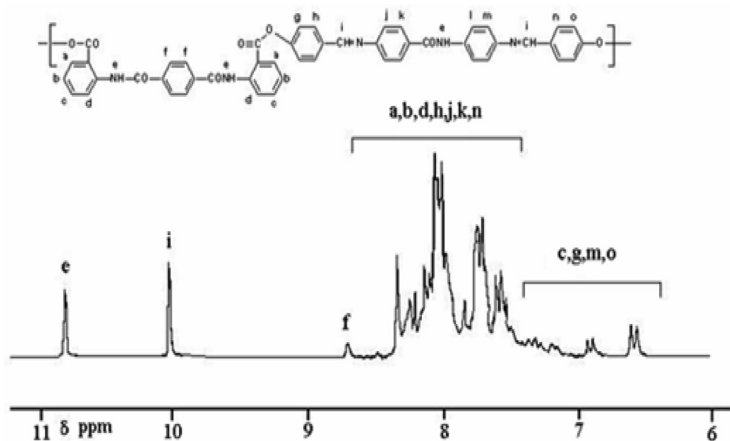


FIGURE 6 $^1\text{H-NMR}$ spectrum of P2C4 in DMSO-d_6 at 200 MHz.

in the range δ 6.59–8.24 ppm. The 8 Ar-H (f,g) for BPA segment resonates at δ 6.59–7.35 ppm, while 4 Ar-H (e) of terephthaloyl group appears at δ 8.14–8.24 ppm as multiplets. The 6 H (a,b,c) of anthranilic acid moiety seems to appear at δ 7.35–8.00 ppm as a multiplet. The singlet of 2 H(d) of two amide groups appears at δ 10.88 ppm. P2C4 (Figure 6): As expected, 4 Ar-H (e) of terephthaloyl segment appears as a multiplet at δ 8.08–8.16 ppm while multiplet in the range δ 6.57–7.38 ppm corresponds to 16 Ar-H (f,g,i,j,k,l) of BHDABA segment [33]. Multiplet in the range δ 7.33–7.99 ppm corresponds to 8 Ar-H (a,b,c) for anthranilic acid moiety. The singlet of 2 H (d) of two amide groups, attached to terephthaloyl group appears at δ 8.28 ppm while singlet of H (k) of amide group of BHDABA moiety seems to appear at δ 8.72 ppm. The sharp singlets at δ 10.04 and 10.78 ppm are attributed to 2 H(h) of two azomethine and 2 H(d) of two amide groups, respectively.

Thermal Studies

The thermograms of P2C1 and P2C4 shown in Figure 7 were obtained in N_2 atmosphere at a heating rate of $10^\circ\text{C}/\text{min}$. The thermogram of P2C1 exhibited thermal stability up to 290°C . The first step degradation continued up to 386°C involving only 16.6% weight loss, with a maximum degradation rate at 335°C . The second step degradation involving about 21% weight loss occurred from 415 to 579°C with maximum rate at 484°C . 51.1% residue was left at 700°C . The first

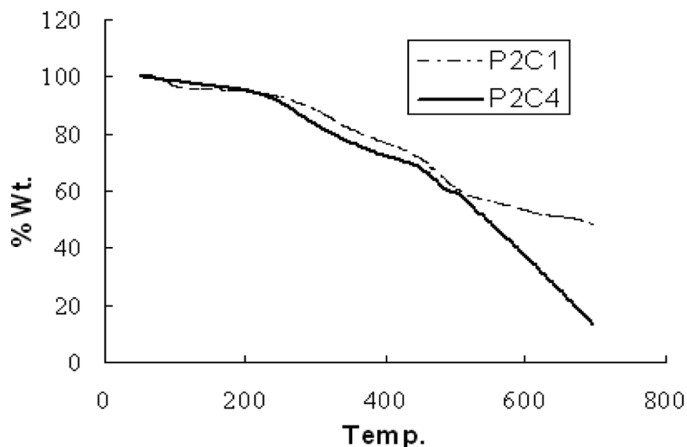


FIGURE 7 TGA of P2C1 and P2C4, in N_2 atmosphere at $10^\circ\text{C}/\text{min}$.

step involving about 28% weight loss for P2C4 commenced at 230°C and extended up to 420°C , with a maximum rate of weight loss at 278°C . The second step degradation occurred over a temperature range 420 to 497°C with T_{max} at 478°C . In the final step from 497 to 700°C , about 45% weight was lost. The temperature at which 50% polyester amide lost was 544°C . At 700°C the residue was about 13.3%.

REFERENCES

- [1] Critchley, J. P., Knight, G. J., and Wright, W. W. (1983). *Heat Resistant Polymers*. Plenum Press, New York.
- [2] Imai, Y., Abe, S., and Ueda, M., *J. Polym. Sci. Polym. Chem. Ed.* **19**, 3285 (1981).
- [3] Kakimoto, M. A., Nagi, Y. S., and Imai, Y., *J. Polym. Sci. Part A Polym. Chem.* **34**, 1511 (1986).
- [4] Sudha, J. D., *J. Polym. Sci. Part A: Polym. Chem.* **38**, 2469 (2000).
- [5] Kalyvas, V., and McIntyre, J. E., *Mol. Cryst. Liq. Cryst.* **80**, 105 (1982).
- [6] Aharoni, S. M., *Macromolecules* **21**, 1941 (1988).
- [7] Gaudiana, R. A., Pickens, N., and Sinta, R., *Mol. Cryst. Liq. Cryst.* **243**, 149 (1994).
- [8] Sudha, J. D., Ramamohan, T. R., Pillai, C. K. S., and Scariah, K. J., *Eur. Polym. J.* **35**, 1637 (1996).
- [9] McCarthy, T. F., Lenz, R. W., Kantor, S. W., and Curran, S., *Macromolecules* **30**, 2825 (1997).
- [10] Sudha, J. D., and Pillai, C. K. S., *J. Polym. Sci. Part A Polym. Chem.* **41**, 335 (2003).
- [11] Carfagna, C., Vacatello, M., and Corradini, P., *J. Polym. Sci. Polym. Chem. Ed.* **15** (1), 1 (1977).
- [12] Schroeder, L. R., and Cooper, S.L., *J. Appl. Phys.* **47** (10), 4310 (1976).
- [13] Goodman, J., *Eur. Polym. J.* **20**, 241 (1984).
- [14] Bozena, K., and Danuta, S., *Polymer.* **36** (26), 5019 (1995).

- [15] Castaldo, L., Candia, F., Maglio, G., Palumbo, R., and Strazza, G., *J. Appl. Polym. Sci.* **27** (5), 1809 (1982).
- [16] Roggero, A. (1983). *Thermotropic Liquid Crystal Polymer Blends*, La Mantia FP. Technomic Publ. Co.
- [17] Grigat, E., Koch, R., and Timmermann, R., *Polym. Degrad. Stab.* **59** (1-3), 223 (1998).
- [18] Thomas, F., Markus, H., Helmut, K., and Hartwig, H., *Polym. Int.* **52**, 1625 (2003).
- [19] Oswal, S. L., and Pandya, A. K., *Iranian Polym. J.* **13** (3), 205 (2004).
- [20] Vogel, A. I. (1956). *A Textbook of Practical Organic Chemistry*. English Language Book Society, 3rd edition London.
- [21] Morgan, P. W. (1965). *Condensation Polymer by Interfacial and Solution Methods*, John Wiley and Sons Inc. New York.
- [22] Cassasa, E. Z., Chao, D. Y., and Henson, M., *J. Macromol. Sci. Chem. A.* **15** (5), 799 (1981).
- [23] Imai, Y., and Tassavori, S., *J. Polym. Sci. Polym. Chem. Ed.* **22**, 1319 (1984).
- [24] Imai, Y., Abe, S., and Ueda, M., *J. Polym. Sci. Polym. Chem. Ed.* **19** (12), 3285 (1981).
- [25] Starks, C. M., *J. Am. Chem. Soc.* **93**, 195 (1971).
- [26] Bilmayer, F. W. Jr. (1994). *Text book of Polymer Science*. John Wiley & Sons, 3rd Ed. New York.
- [27] Patil, A. O., Deshpande, D. D., Talwar, S. S., and Biswas, A. B., *J. Polym. Sci. Polym. Chem. Ed.* **19**, 1155 (1981).
- [28] Jeong, H. J., Kakimoto, M. A., and Imai, Y., *J. Polym. Sci. Part A Polym. Chem.* **29** (9), 1293 (1991).
- [29] Idage, S. B., Idage, B. B., and Vernekar, S. P., *J. Polym. Sci. Part A Polym. Chem.* **30**, 513 (1992).
- [30] Desai, D. R. (1995). Synthesis and studies of physical, spectral and thermal properties of homo-, co-polycyanurates. Ph. D. Thesis, South Gujarat University, Surat.
- [31] Kim, S. G., Han, Y. K., and Gong, M. S., *Bull. Korean Chem. Soc.* **16** (4), 326 (1995).
- [32] Rao, M. V., Rajivadiya, A. J., Parsania, P. H., and Parekh, H. H., *J. Macromol. Sci. Chem.* **A27**, 53 (1990).
- [33] Mahajan, S. S., Idage, B. B., Chavan, N. N., and Sivaram, S., *J. Appl. Polym. Sci.* **61** (13), 2297 (1996).