

Synthesis and Properties of Polyamides Based on 1,1-Bis[4-(4-aminophenoxy)phenyl]cyclohexane

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ABSTRACT: An ether diamine monomer containing the cyclohexane structure, 1,1-bis[4-(4-aminophenoxy)phenyl]cyclohexane (BAPC), was prepared through nucleophilic substitution reaction of 1,1-bis(4-hydroxyphenyl)cyclohexane and *p*-chloronitrobenzene in the presence of K_2CO_3 , followed by catalytic hydrazine reduction. Using triphenyl phosphite and pyridine as condensing agent, novel polyamides were directly polycondensated from BAPC with various aromatic dicarboxylic acids in the *N*-methyl-2-pyrrolidone (NMP) solution containing dissolved calcium chloride. The resulting polyamides had inherent viscosities from 0.97 to 1.61 dL g⁻¹. Most had good solubility in polar organic solvents such as NMP and *N,N*-dimethylacetamide, and afforded transparent, flexible, and tough films by solution casting. The glass transition temperatures (T_g) of the polymers were 188–240°C and 10% mass loss temperatures were recorded at 473–499°C in air. A comparative study on the properties with corresponding polyamides derived from 1,1-bis[4-(4-carboxyphenoxy)phenyl]cyclohexane (BCPC) was made.

KEY WORDS Polyamides / 1,1-Bis[4-(4-aminophenoxy)phenyl]cyclohexane / Triphenyl Phosphite /

Wholly aromatic polyamides belong to the class of high-performance polymers due to their high thermal and chemical resistance as well as high modulus.^{1,2} Kevlar® [poly(*p*-phenylene)terephthalamide] and Nomex® [poly(*m*-phenylene)isophthalamide] are commercially marketed as high-performance polymers usually in fiber form. Both polyamides exhibit excellent mechanical strength and thermal stability and are suitable for composites. Versatile applications of these polyamides have generally been reduced by the high crystallinity and limited solubility in common solvents.

Therefore, the preparation of soluble and/or thermoplastic polyamides has been a major research interest. To accomplish this goal, many attempts have been made with the aim of synthetic modification of the rigid backbone by the incorporation of flexible bridging linkages,^{3–5} distortion of molecular symmetry by *meta*- or *ortho*-catenated aromatic units^{6–10} and introduction of bulky groups into the polymer chain.^{11–24} The latter is probably the most common approach. In addition to bulky nonlinear monomers, any of these procedures can also be used to disrupt crystallinity to obtain almost completely amorphous polyamides. In general, amorphous polyamides have relatively low softening temperatures and enhanced solubility compared to crystalline analogs, and are also of considerable interest as high-performance engineering plastics.

An alternative way to achieve the above objectives is to use cardo monomers such as fluorene, phthalide, and phthalimidine groups.^{25–28} This approach may improve solubility while maintaining high T_g through controlled segmental mobility. Although some polyimides and polyamides of 1,1-bis[4-(4-aminophenoxy)phenyl]cyclohexane (BAPC) are known,^{29,30} to our knowledge, little is known about BAPC polymers. This article deals with the synthesis and basic characterization of polyamides (designated as the PA-I series as shown in Scheme

1) based on BAPC and various aromatic dicarboxylic acids. For a comparative study, analogous polyamides (PA-II) based on 1,1-bis[4-(4-carboxyphenoxy)phenyl]cyclohexane (BCPC) and aromatic diamines were synthesized and characterized.

EXPERIMENTAL

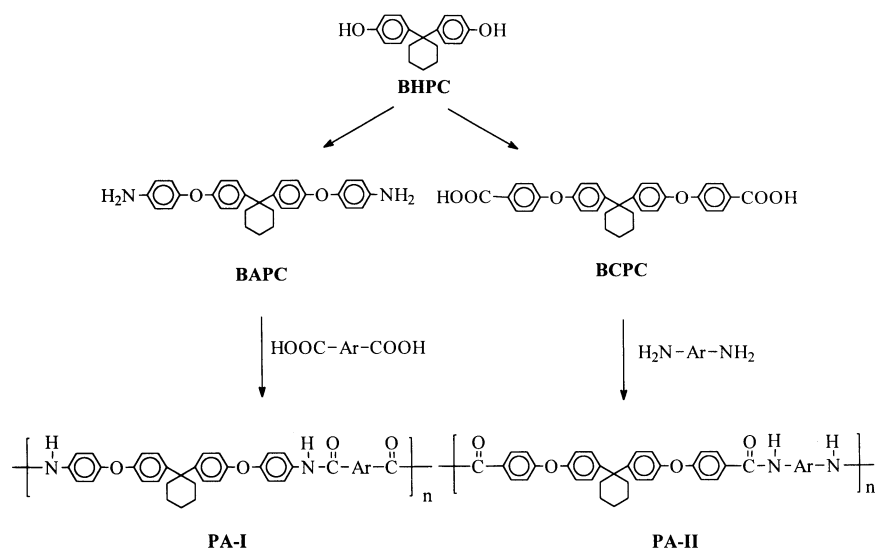
Materials

1,1-Bis(4-hydroxyphenyl)cyclohexane (BHPC, from TCI), *p*-chloronitrobenzene (from TCI), hydrazine monohydrate (from Wako), 10 % palladium on activated carbon (from Fluka) were used as received. *N,N*-Dimethylformamide (DMF, from Fluka), *N*-methyl-2-pyrrolidone (NMP, from Fluka), *N,N*-dimethylacetamide (DMAc, from Fluka), and pyridine (Py, from Wako) were purified by distillation under reduced pressure over calcium hydride and stored over 4 Å molecular sieves. Triphenyl phosphite (TPP, from TCI) was purified by vacuum-distillation. Commercially obtained calcium chloride (from Wako) was dried under vacuum at 180°C for 10 h. Reagent-grade aromatic dicarboxylic acids, such as terephthalic acid (III_a, Fluka), isophthalic acid (III_b, Wako), 4,4'-biphenyldicarboxylic acid (III_c, TCI), 4,4'-oxydibenzoic acid (III_d, TCI), 5-*t*-butylisophthalic acid (III_e, Amoco Chemical Co.), 2,6-naphthalenedicarboxylic acid (III_f, TCI), 4,4'-sulfonyldibenzoic acid (III_g, New Japan Chemical Co.), and 2,2-bis(4-carboxyphenyl)-1,1,1,3,3,3-hexafluoropropane (III_h, Chriskev) were used as received. BCPC was synthesized from the fluoro-displacement of *p*-fluorobenzonitrile with BHPC in the presence of potassium carbonate, followed by alkaline hydrolysis. Details of the synthesis and characterization data of BCPC have been reported previously.³¹

Monomer Synthesis

1,1-Bis[4-(4-nitrophenoxy)phenyl]cyclohexane (BNPC). A mixture of 26.8 g (0.1 mmol) of BHPC, 32.3 g (0.205

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Scheme 1.

mmol) of *p*-chloronitrobenzene, and 28 g (0.2 mmol) of anhydrous K_2CO_3 was refluxed at 150–160°C in 120 mL DMF for 8 h. The mixture was cooled to room temperature and poured into 200 mL methanol to precipitate a yellow solid which was collected by filtration, washed with hot water, and dried. The yield of the product was 50 g. The crude product was purified by recrystallization from DMF/methanol. The yield of the purified product was 47.7 g (94%); mp 127–128°C (lit.³⁰ 133.8–134.1°C).

IR (KBr): 1516 and 1341 ($-\text{NO}_2$), 1238 cm^{-1} ($-\text{O}-$).

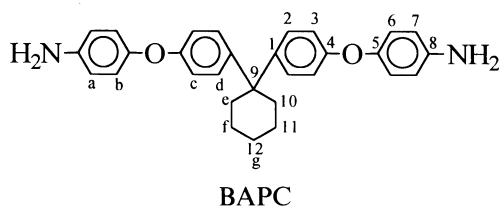
Anal. Calcd for $\text{C}_{30}\text{H}_{26}\text{N}_2\text{O}_6$ (510.55): C, 70.58%; H, 5.13%; N, 5.49%. Found: C, 70.73%; H, 5.10%; N, 5.57%.

1,1-Bis[4-(4-aminophenoxy)phenyl]cyclohexane (BAPC). 20.4 g (0.04 mmol) of the dinitro compound BNPC, 0.15 g of 10% Pd/C, and 100 mL ethanol were placed in a 250-mL three-necked flask. Hydrazine monohydrate (12 mL) was added dropwise to the mixture at 80°C during 1 h. The system was refluxed overnight (about 8 h) and filtered hot to remove the catalyst. Upon cooling, white crystals precipitated (BAPC), and were isolated by filtration and dried in vacuum. Yield: 17.8 g (99%); mp 156–157°C (lit.³⁰ 155.0–155.5°C).

IR (KBr): 3360 (N–H str.), 2932 (aliphatic C–H str.), 1231 cm^{-1} (C–O str.).

^1H NMR dimethyl sulfoxide- d_6 (DMSO- d_6): δ 7.17 (d, H_d , 4H), 6.72 (two overlapped AB doublets, H_{b+c} , 8H), 6.57 (d, H_a , 4H), 4.92 (s, $-\text{NH}_2$, 4H), 2.17 (s, H_e , 4H), 1.42 ppm (s, H_{f+g} , 6H).

^{13}C NMR (DMSO- d_6): 156.38 (C^4), 145.52, 145.30 ($\text{C}^5 + \text{C}^8$), 141.59 (C^1), 127.76 (C^2), 120.85 (C^6), 115.90 (C^3), 114.76 (C^7), 44.31 (C^9), 36.28 (C^{10}), 25.66 (C^{11}), 22.46 ppm (C^{12}).



Anal. Calcd for $\text{C}_{30}\text{H}_{30}\text{N}_2\text{O}_2$ (450.58): C, 79.97%; H, 6.71%; N, 6.22%. Found: C, 79.91%; H, 6.72%; N, 6.25%.

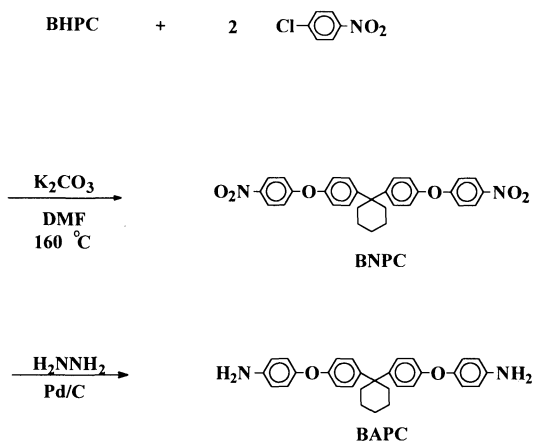
Polymerization

Polyamides were prepared by direct polycondensation as follows. A mixture of 0.45 g (1.0 mmol) of diamine BAPC, 0.166 g (1.0 mmol) of terephthalic acid (**III_a**), 0.15 g of calcium chloride, 0.17 g of lithium chloride, 0.6 mL of pyridine, 0.6 mL of triphenyl phosphite, and 2 mL of NMP was heated with stirring at 100°C. An additional 1.5 mL NMP were added to the reaction mixture as the solution became too viscous to stir. After 3 h stirring at 100°C, it was slowly poured into 100 mL of stirred methanol giving rise to a stringy precipitate which was washed thoroughly with methanol and hot water, collected by filtration and dried at 100°C under vacuum. The yield was 0.61 g (99%). The inherent viscosity of the polyamide **PA-I_a** was 1.03 dL g^{-1} , as measured at a concentration of 0.5 g dL^{-1} in DMAc at 30°C.

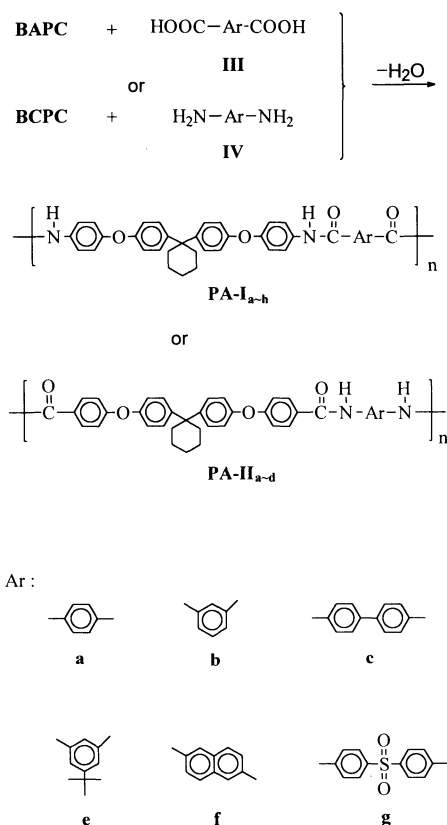
IR (film): 3318 (N–H str.), 1657 (C=O str.), 1235 cm^{-1} (C–O–C str.).

Measurements

Elemental analysis was run in a Perkin-Elmer Model 2400 C, H, N, analyzer. IR spectra were recorded on a Jasco FT/IR-7000 Fourier-transform Infrared Spectrometer. ^{13}C and ^1H NMR spectra were obtained on a Jeol EX-400 NMR spectrometer with perdeuteriodimethyl sulfoxide (DMSO- d_6) and tetramethylsilane (TMS), respectively, as an internal reference. Inherent viscosity was measured with a Cannon-Fenske viscometer at 30°C. DSC traces were measured on a TA Instrument DSC 2010 at the rate of 15°C min^{-1} in a nitrogen flow (40 $\text{cm}^3 \text{min}^{-1}$). Thermogravimetry (TG) analysis was conducted with a Rigaku thermoflex TG 8110 coupled to a Rigaku TAS-100 thermal analysis station. Experiments were carried out on 10 ± 2 mg samples heated in flowing nitrogen (50 $\text{cm}^3 \text{min}^{-1}$) at a heating rate of 20°C min^{-1} . An Instron universal tester model 1130 with a load cell of 5 kg was used to study



Scheme 2.



Scheme 3.

the stress-strain behavior of the samples. A gauge length of 2 cm and strain rate of 5 cm min⁻¹ were used for this study. Measurements were performed at room temperature with film specimens (0.5 cm wide, 6 cm long, and 0.05 mm thick), and the average of at least five individual determinations was used.

RESULTS AND DISCUSSION

Monomer Synthesis

Bis(ether amine) monomer BAPC containing the 1,1-cyclohexylidene group was prepared in two steps according to Scheme 2. The intermediate dinitro compound BNPC was synthesized by nucleophilic aromatic substitution of bisphenol BHPC with *p*-chloronitrobenzene in the presence of anhydrous potassium carbonate in DMF. The diamine BAPC was obtained in high pu-

Table I. Synthesis conditions and inherent viscosities of polyamides^a

PA-	Amounts of reagents used ^b					η_{inh}^c dL g ⁻¹
	NMP mL	Py mL	TPP mL	CaCl ₂ g		
I _a	2.2+1.5 ^d	0.6	0.6	0.15 (+0.17 g LiCl)	1.03	
I _b	2+2	0.6	0.6	0.1 (+ 0.1 g LiCl)	1.37	
I _c	3	1	0.6	0.2	1.33	
I _d	2.5+1	0.8	0.6	0.2	1.61	
I _e	2	0.6	0.6	0.2	0.97	
I _f	2.5+2	0.8	0.6	0.2	1.40	
I _g	2.2	0.7	0.6	0.17	1.07	
I _h	2.5+1	0.8	0.6	0.17	1.15	
II _a	3	1	0.9	0.4	0.75	
II _b	3	1	0.9	0.4	0.89	
II _c	4+3	1	0.9	0.5	1.78	
II _d	3+1.5	1	0.9	0.4	1.61	

^a 1.0 mmol (for PA-I) and 1.5 mmol (for PA-II) each of the diacid and diamine monomer were used. ^b NMP, *N*-methyl-2-pyrrolidone; Py, pyridine; TPP, triphenyl phosphite. ^c Measured in DMAc at 30 °C on 0.5 g dL⁻¹. ^d "2.2+1.5" means that an initial amount of 2.2 mL of NMP was used and an additional 1.5 mL NMP were added when the reaction solution was too viscous to stir.

rity and high yield using hydrazine hydrate and Pd/C catalyst in refluxing ethanol. The structures of BNPC and BAPC were confirmed by FT-IR, NMR, and elemental analysis.

Polymer Synthesis

Polyamides PA-I_{a-h} were prepared using the Yamazaki³² phosphorylation polycondensation procedure from the diamine BAPC and dicarboxylic acids III_{a-h} (Scheme 3). Synthesis conditions and inherent viscosities of the polyamides are summarized in Table I. All polymerizations proceeded homogeneously throughout the reaction and highly viscous solutions were obtained. The polyamides were isolated as tough fibers in quantitative yields. Inherent viscosities of PA-I_{a-h} ranged from 0.97 to 1.61 dL g⁻¹, indicating high molecular weight polymers. For comparison, synthesis conditions and inherent viscosities of some analogous polyamides PA-II_{a-d} are listed in Table I.

The results of elemental analyses of the polyamides are listed in Table II. In most cases, the values for carbon and nitrogen were lower than those calculated for the proposed structures, but hydrogen was higher, possibly due to the hygroscopic nature of the samples caused by the amide groups of the polymers. Saturated moisture absorption was 1.34–3.54 wt% at room temperature and normal pressure at this laboratory. Corrected values were in agreement with those calculated after absorbed moisture was excluded.

Polymer Properties

The solubility behavior of the polyamides is shown in Table III. All polyamides of PA-I exhibited excellent solubility in NMP and DMAc. Except for polymer (PA-I_c) derived from benzidine, all PA-I polymers were soluble in DMF, DMSO, *m*-cresol, and pyridine. Polyamides I_g and I_h were also soluble in tetrahydro-

Table II. Elemental analysis of polyamides

PA-	Formula M_w		Elemental analysis ^b /%			Moisture intake ^a /%
			C	H	N	
I_a	(C ₃₈ H ₃₂ O ₄ N ₂) _n (580.68) _n	Calcd	78.60	5.55	4.82	3.32
		Found	75.99	5.59	4.73	
		Corrected	78.51	5.40	4.89	
I_b	(C ₃₈ H ₃₂ O ₄ N ₂) _n (580.68) _n	Calcd	78.60	5.55	4.82	3.41
		Found	75.92	5.57	4.85	
		Corrected	78.51	5.38	5.02	
I_c	(C ₄₄ H ₃₆ O ₄ N ₂) _n (656.78) _n	Calcd	80.47	5.52	4.27	3.54
		Found	77.62	5.56	4.21	
		Corrected	80.36	5.08	5.28	
I_d	(C ₄₄ H ₃₆ O ₄ N ₂) _n (672.78) _n	Calcd	78.55	5.39	4.16	3.04
		Found	76.15	5.41	4.09	
		Corrected	78.46	5.24	4.21	
I_e	(C ₄₂ H ₄₁ O ₄ N ₂) _n (637.80) _n	Calcd	79.09	6.48	4.39	2.54
		Found	77.08	6.33	4.42	
		Corrected	79.04	6.17	4.53	
I_f	(C ₄₂ H ₃₄ O ₄ N ₂) _n (630.74) _n	Calcd	79.98	5.43	4.44	2.83
		Found	77.72	5.49	4.35	
		Corrected	79.92	5.33	4.47	
I_g	(C ₄₄ H ₃₆ O ₄ N ₂ S ₁) _n (720.84) _n	Calcd	73.32	5.03	3.89	3.22
		Found	70.96	5.09	3.83	
		Corrected	73.24	4.93	3.95	
I_h	(C ₄₇ H ₃₆ O ₄ N ₂ F ₆) _n (806.80) _n	Calcd	69.97	4.50	3.47	1.34
		Found	69.03	4.59	3.52	
		Corrected	69.96	4.53	3.57	

^a Moisture intake (%) = $[(W - W_0)/W_0] \times 100\%$; W = constant weight of the polymer sample after standing at room temperature, and W_0 = weight of the polymer sample after being 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 %).

Table III. Solubility behavior of polyamides^a

PA-	Solvent ^b						
	NMP	DMAc	DMF	DMSO	<i>m</i> -Cresol	Py	THF
I_a	+	+	+	+	+	+	-
I_b	+	+	+	+	+	+	S
I_c	+	+	S	S	-	-	-
I_d	+	+	+	+	+	+	S
I_e	+	+	+	+h	+	+	-
I_f	+	+	+	+	+	+	-
I_g	+	+	+	+	+	+	+
I_h	+	+	+	+	+	+	+
II_a	+	+	+	+h	+h	S	-
II_b	+	+	+	+	+	+	-
II_c	+	+h	-	-	-	-	S
II_d	+	+	+	+	+h	+	-

^a +, soluble at room temperature; S, swelling; +h, soluble on heating; -, insoluble even on heating. ^b NMP, *N*-methyl-2-pyrrolidone; DMAc, *N,N*-dimethylacetamide; DMF, *N,N*-dimethylformamide; DMSO, dimethyl sulfoxide; Py, pyridine.

furan due to the presence of sulfonyl and hexafluoroisopropylidene groups. The lesser solubility of polyamide **PA-I_c** may indicate strong inter-chain interaction or good packing ability. In comparison with some analogous polyamides with an isomeric repeating unit, **PA-I_{a-d}** and **PA-II_{a-d}** revealed similar solubility behavior.

X-Ray diffraction patterns showed that polyamides **I_{a-h}** are all amorphous. One factor contributing to the improved solubility was the characteristic amorphous

Table IV. Tensile properties of polyamide films^a

PA-	Strength at yield	Strength at break	Elongation to break	Initial modulus
	MPa	MPa	%	GPa
I_a	93	88	28	2.09
I_b	—	97	7	2.02
I_c	—	77	8	2.14
I_d	85	79	46	1.70
I_e	—	88	8	2.03
I_f	—	84	16	1.80
I_g	—	94	12	2.30
I_h	—	77	15	1.82

^a Films were cast from slow evaporating polymer solutions in DMAc.

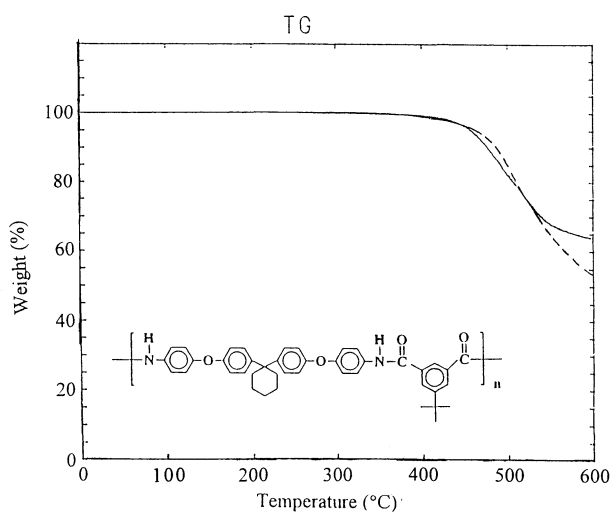


Figure 1. TG curves for **PA-I_e** in nitrogen (—) and in air (---) with a heating rate of 20°C min⁻¹.

nature induced by the introduction of the cyclohexylidene cardo group and flexible ether linkage. All polyamides afforded tough and flexible films by solution casting. Table IV summarizes the tensile properties of the polyamide films. **PA-I** polyamides had tensile strength of 77–97 MPa, elongation at break of 7–46%, and initial modulus of 1.70–2.30 GPa. Polyamides **I_a** and **I_d** showed clear yield points in the stress-strain curves and had moderate elongation at break, indicating high toughness.

The thermal properties of all the polyamides were evaluated by TG analysis and differential scanning calorimetry (DSC). Figure 1 shows typical TG thermograms for the representative polyamide **I_e** in air and nitrogen. Polyamide **I_e** started to lose weight around 450°C. In nitrogen, this polymer left 64% char residue when heated to 600°C. All the polyamides showed similar decomposition with no significant weight loss below 450°C in nitrogen or air. The data in Table V show that the 10% weight loss temperatures (T_d) of the **PA-I** series were 463–485°C in nitrogen and 472–498°C in air, and char yields at 600°C in nitrogen were all higher than 51%. It is interesting to note that in all cases the temperature at which 10% weight loss occurred in air was higher than that in nitrogen. This may be attributable to an early weight-gained oxidation and possible oxidative interchain crosslinking of the aliphatic seg-

Table V. Thermal properties of polyamides

PA-	T_g^a	$T_d^{b/c}$		Char yield ^d
	°C	In N ₂	In air	
I _a	202	470	480	62
I _b	188	483	492	59
I _c	240	485	486	55
I _d	194	474	488	59
I _e	234	477	487	64
I _f	217	469	481	58
I _g	224	463	473	51
I _h	213	483	499	56
II _a	205	470	481	59 ^d
II _b	183	472	490	46 ^d
II _c	245	460	473	55 ^d
II _d	195	485	494	53 ^d

^a Baseline shift in second heating DSC traces, at 15°C min⁻¹. ^b Decomposition temperatures at which 10% weight loss was recorded by TG at a heating rate 20°C min⁻¹. ^c Residual weight (%) when heated to 600°C in nitrogen. ^d Residual weight (%) when heated to 800°C in nitrogen.

ments in the polymer chain when thermally degraded in air.

DSC measurements were conducted at a heating rate of 15°C min⁻¹ in nitrogen. Quenching from the elevated temperatures (approximately 400°C) to room temperature in air gave predominantly amorphous samples so that the glass transition temperatures (T_g) of all polyamides could be observed clearly in the second heating traces of DSC. Polyamides I_{a-h} exhibited T_g between 188 and 240°C, following decreasing order of chain flexibility and steric hindrance of the dicarboxylic acids. The lowest T_g of 188°C was for polyamide I_b derived from isophthalic acid. A 46°C difference in T_g between PA-I_b and I_e was observed as a result of a greater rotational barrier due to the *t*-butyl group of diacid III_e. Polyamide I_d showed a relatively lower T_g of 194°C due to the presence of ether linkages in the diacid component. In comparison with polyamides with the isomeric recurring unit, PA-I_{a-d} displayed a comparable T_g with corresponding PA-II_{a-d}, thus indicating that the presence of cyclohexylidene in either diamine or dicarboxylic acid moiety did not affect noticeably polymer rigidity.

CONCLUSIONS

It is concluded that the introduction of the cyclohexylidene cardo group and ether linkages into the backbones of aromatic polyamides improves solubility with lowered T_g s compared to wholly aromatic polyamides. Improved solubility and decreased T_g were due to the amorphous nature of the polyamides that had resulted from flexible kinks and the plasticizing effect of the cyclohexylidene unit. The polyamides showed reasonable thermal stability and a large window between decomposition and glass transition temperatures, which are advantageous in the processing of these polymers.

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