

Synthesis and Characterization of Organosoluble Poly(arylate-imide)s Prepared from Direct Polycondensation of Bis(trimellitimide)-Diacids and Various Bisphenols

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ABSTRACT: Three diimide-diacids, 2,2-bis[4-(4-trimellitimidophenoxy)phenyl]hexafluoropropane (**I-A**), 2,2-bis[4-(4-trimellitimidophenoxy)phenyl]propane (**I-B**), and 5,5'-bis[4-(4-trimellitimidophenoxy)phenyl]hexahydro-4,7-methanoindan (**I-C**), were prepared by the azeotropic condensation of trimellitic anhydride with three analogous diamines. Three series of alternating aromatic poly(arylate-imide)s, having inherent viscosities of 0.41–0.82 dL/g, were synthesized from these diimide-diacids (**I-A**, **I-B**, and **I-C**) with various bisphenols by direct polycondensation using diphenyl chlorophosphate and pyridine as condensing agents. All of the polymers were readily soluble in a variety of organic solvents such as *N*-methyl-2-pyrrolidone, *N,N*-dimethylacet-

amide, and even in the less polar tetrahydrofuran. These polymers could be cast into transparent and tough films, which had strength at break values ranging from 73 to 98 MPa, elongation at break from 6 to 11%, and initial modulus from 1.6 to 2.2 GPa. The softening temperatures of the polymers were recorded at 145–248°C. They had 10% weight loss at a temperature above 450°C and left 35–51% residue even at 800°C in nitrogen. © 2003 Wiley Periodicals, Inc. *J Appl Polym Sci* 89: 3818–3825, 2003

Key words: direct polycondensation; organosoluble; poly(arylate-imide)s; bis(trimellitimide)-diacids

INTRODUCTION

Wholly aromatic polyimides are widely used in the semiconductor and electronic packaging industry because of their outstanding thermal stability, high mechanical strength, good insulation properties with low dielectric constants, good adhesion to common substrates, and superior chemical stability.^{1,2} However, these polymers are generally intractable and lack the properties essential for successful fabrication into useful forms because of their high melting or glass-transition temperatures and limited solubility in organic solvents. Therefore, copolyimides, such as poly(amide-imide)s and poly(arylate-imide)s, have been developed.

Aromatic poly(arylate-imide)s can be synthesized from three kinds of condensing monomers in three groups: trimellitic anhydride (TMA), aromatic diamine, and bisphenol; aromatic aminophenol, dianhydride, and aromatic diacid; and aromatic amino acids, dianhydride, and bisphenol. The five general methods

used in the synthesis of regular poly(arylate-imide)s are polycondensation of an ester-group-containing dianhydride with a diamine,^{3–6} polycondensation of an ester-group-containing diamine with a dianhydride,⁷ a reaction between a dicarboxylic acid containing an imide ring with a bisphenol,^{8–13} a reaction between a bisphenol containing an imide ring with a dicarboxylic acid, and a self-reaction of an imide-containing phenol-acid monomer.^{14–16} The first two methods proceed through the solution polycondensation of polyimide, and the other three methods proceed through the melting transesterification of an imide-group-containing monomer or the solution ester polycondensation of its derivative. There is usually difficulty in stirring in the process of melting transesterification, but solution polycondensation goes smoothly because it is easy to stir. However, there are few common solvents for dissolving imide and arylate groups. The unsatisfactory results obtained from solution polycondensation are caused by the poor solubility of these polymers, resulting in early precipitation within the reaction medium and retarding further polymerization. These poly(arylate-imide)s also have relatively low inherent viscosities.

In the present study, three diimide-dicarboxylic acids^{17–19} are prepared from 2,2-bis[4-(4-aminophenoxy)phenyl]hexafluoropropane (6FBAPP), 2,2-bis[4-(4-

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aminophenoxy)phenyl]propane (BAPP), and 5,5'-bis[4-(4-aminophenoxy)phenyl]-4,7-methano-hexahydroindan (BAMI) with TMA. These diimide-diacids and several bisphenols, which remain homogeneous during the reaction in pyridine (Py), are used to prepare novel poly(arylate-imide)s that have higher degrees of polymerization and are soluble in amide-type solvents. These polymerizations avoid the use of moisture-sensitive acid chlorides and they can proceed directly using diphenyl chlorophosphate (DPCP) as a condensing agent. The solubility, tensile properties, and thermal properties of the obtained polymers were also investigated.

EXPERIMENTAL

Materials

The TMA (Wako), Py (Wako), and DPCP (Merck) used in the study were used as received. The diamines 6FBAPP and BAPP were obtained from Chriskev Corp. and used as received. BAMI was prepared according to the method reported in the preceding paper.¹⁹ The bisphenols, including 2,2'-bis(4-hydroxyphenyl)propane (**II_a**, Wako), 2,2'-bis(4-hydroxyphenyl)hexafluoropropane (**II_b**, Asahi Glass Co.), 2,2',6,6'-tetramethylbiphenol (**II_c**, Mitsubishi), 1,1-bis(4-hydroxyphenyl)-1-phenylethane (**II_d**, Honshu Chem. Co.), 5,5'-bis(4-hydroxyphenyl)hexahydro-4,7-methanoindan (**II_e**, Wako), phenolphthalein (**II_f**, Wako), and 4,4'-(1,4-phenylenediisopropylidene)diphenol (**II_g**, Mitsui), were used without further purification. Commercially obtained anhydrous lithium chloride (LiCl) was dried under a vacuum at 150°C for 10 h.

Synthesis of diimide-diacids I-A-C

The synthesis of the diimide-diacids was based on a procedure reported elsewhere,¹⁷⁻²⁰ and an example of this method for **I-A** is as follows. A mixture of 8.67 g (10 mmol) of 6FBAPP and 10.65 g (20 mmol) of TMA was heated in 45 mL of dry *N*-methyl-2-pyrrolidone (NMP) at 60°C for 1 h. About 20 mL of toluene was then added, and the mixture was heated at reflux for 3 h until about 0.35 mL of water was distilled off azeotropically via a Dean-Stark trap. Heating was continued to distill off the residual toluene. After cooling, about 30 mL of methanol was added and the precipitated product was isolated by filtration, purified by recrystallization from DMF, and dried in a vacuum to give 17.89 g of **I-A** (yield 92.6%, mp 337°C). FTIR (KBr) ν_{\max} (cm⁻¹): 3465 (acid —OH), 1782 (imide, symmetric C=O stretching), 1722 (acid C=O stretching and asymmetric imide C=O stretching), 1390 (imide, imide ring vibration, axial), 1117 (imide, imide ring vibration, transverse), 728 (imide, imide ring vibration, out of plane). ELEM. ANAL. Calcd for

C₄₅H₂₄O₁₀N₂F₆: C, 62.35%; H, 2.79%; N, 3.23%. Found: C, 62.22%; H, 2.91%; N, 3.05%.

Other diimide-diacids were prepared in an analogous manner.

Polymerization of poly(arylate-imide)s

A typical example of polycondensation is now described. A solution of DPCP (0.6 mL), LiCl (0.05 g), and Py (4.0 mL) was stirred at room temperature for 30 min and then added dropwise over 20 min to a hot solution (preheated at 120°C for 5 min) containing 0.866 g (1 mmol) of diimide-diacid **I-A** and 0.226 g (1 mmol) of bisphenol **II_a** in Py (2.0 mL). The final solution was heated at 120°C for 3 h under stirring. The obtained polymer solution was trickled into 200 mL of 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 a vacuum. The yield was quantitative. The inherent viscosity of the polymer **III-A_a** in *N,N*-dimethylacetamide (DMAc) at a 0.5 g/dL concentration at 30°C was 0.51 dL/g. FTIR (KBr) ν_{\max} (cm⁻¹): 1782, 1722 (symmetric and asymmetric imide C=O stretching), 1245 (C—O—C). ¹H-NMR (400 MHz, CDCl₃, δ): 8.745 (s, 2H, H_a), 8.626 (d, 2H, H_b), 8.093 (d, 2H, H_c), 7.473 (d, 4H, H_d), 7.409 (d, 4H, H_e), 7.348 (d, 4H, H_f), 7.196 (dd, 8H, H_i+H_j), 7.063 (d, 4H, H_e), 1.845 (s, 6H, H_g). ¹³C-NMR (100 MHz, CDCl₃, δ): 166.24 (C²); 163.30 (C¹); 157.45 (C¹³), 156.04 (C¹²); 148.51 (C¹⁹); 136.35 (C²²); 135.62, 135.46 (C^{6,16}); 132.08, 131.89 (C^{3,8}); 129.86 (C⁵); 128.11, 127.92 (C^{15,21}); 127.02 (C⁹); 125.34 (C⁴); 124.21 (¹J_{CF}, quartet, with ¹J_{CF} = 280 Hz); 124.04 (C⁷); 123.43 (C¹⁰); 120.88 (C²⁰); 120.15, 119.97 (C^{10,14}); 118.21 (C¹¹); 63.83 (²J_{CF}, triplet, with ²J_{CF} = 25 Hz); 64.55 (C²³); 30.98 (C²⁴).

Other poly(arylate-imide)s were synthesized in an similar manner. Representative IR spectra and elemental analysis results are shown in Figure 1 and Table I.

Preparation of poly(arylate-imide) films

An approximately 10% polymer solution was made by dissolving poly(arylate-imide) in DMAc. The solution was poured into a 9-cm diameter glass culture dish that was placed in a 90°C oven overnight to remove the solvent. Then the obtained semidried polymer film was stripped from the glass substrate and further dried in a vacuum at 160°C for 6 h. The obtained films were about 0.05 mm thick.

Measurements

The ¹H- and ¹³C-NMR spectra were determined on a Jeol EX-400 FT-NMR spectrometer, and the IR spectra were recorded on a Horiba FTIR-720 Fourier trans-

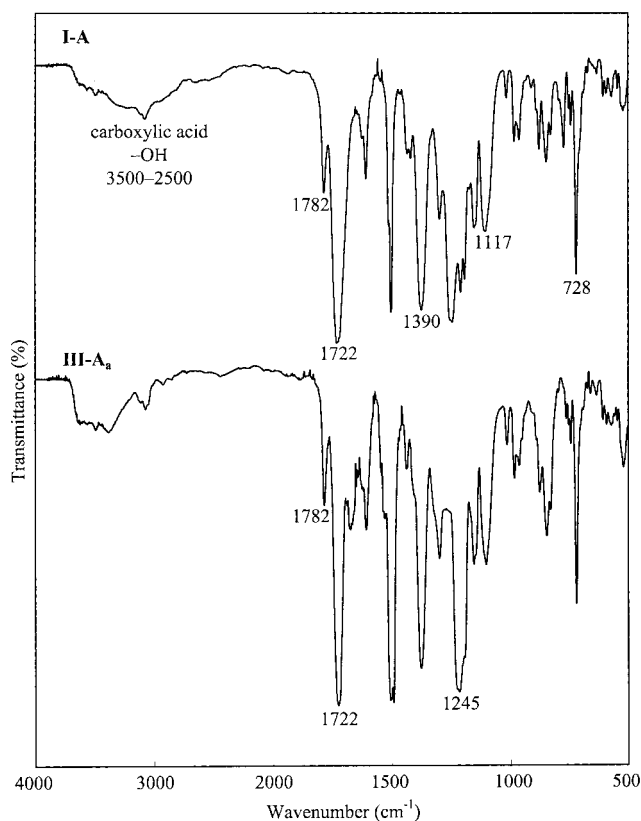


Figure 1 The FTIR spectra of diimide-diacid I-A and poly(arylate-imide) III-A_a.

form IR spectrometer. Elemental analyses were carried out with a Perkin–Elmer model 2400 C, H, N analyzer. The inherent viscosities of all polymers were

determined using a Cannon–Fenske viscometer. Thermomechanical analyses (TMA) were conducted with a Perkin–Elmer TMA 7 instrument from 40 to 300°C at a scan rate of 10°C/min using a 1.0-mm diameter penetration probe under an applied constant load of 10 mN. The softening temperatures were taken as the onset temperature of probe displacement on the TMA traces. Thermogravimetry analyses (TGA) were conducted with a TA Instruments TGA 2050. Experiments were carried out on 10 ± 2 mg samples heated in flowing nitrogen or air ($100 \text{ cm}^3/\text{min}$) at a heating rate of 20°C/min. An Instron universal tester (model 1130) with a load cell of 5 kg was used to study the stress–strain behavior of the samples. A gauge of 2 cm and a strain rate of 5 cm/min were used for the study. Measurements were performed at room temperature with film specimens (0.5 cm wide, 6 cm long, and about 0.05 mm thick), and an average of at least five individual determinations was reported.

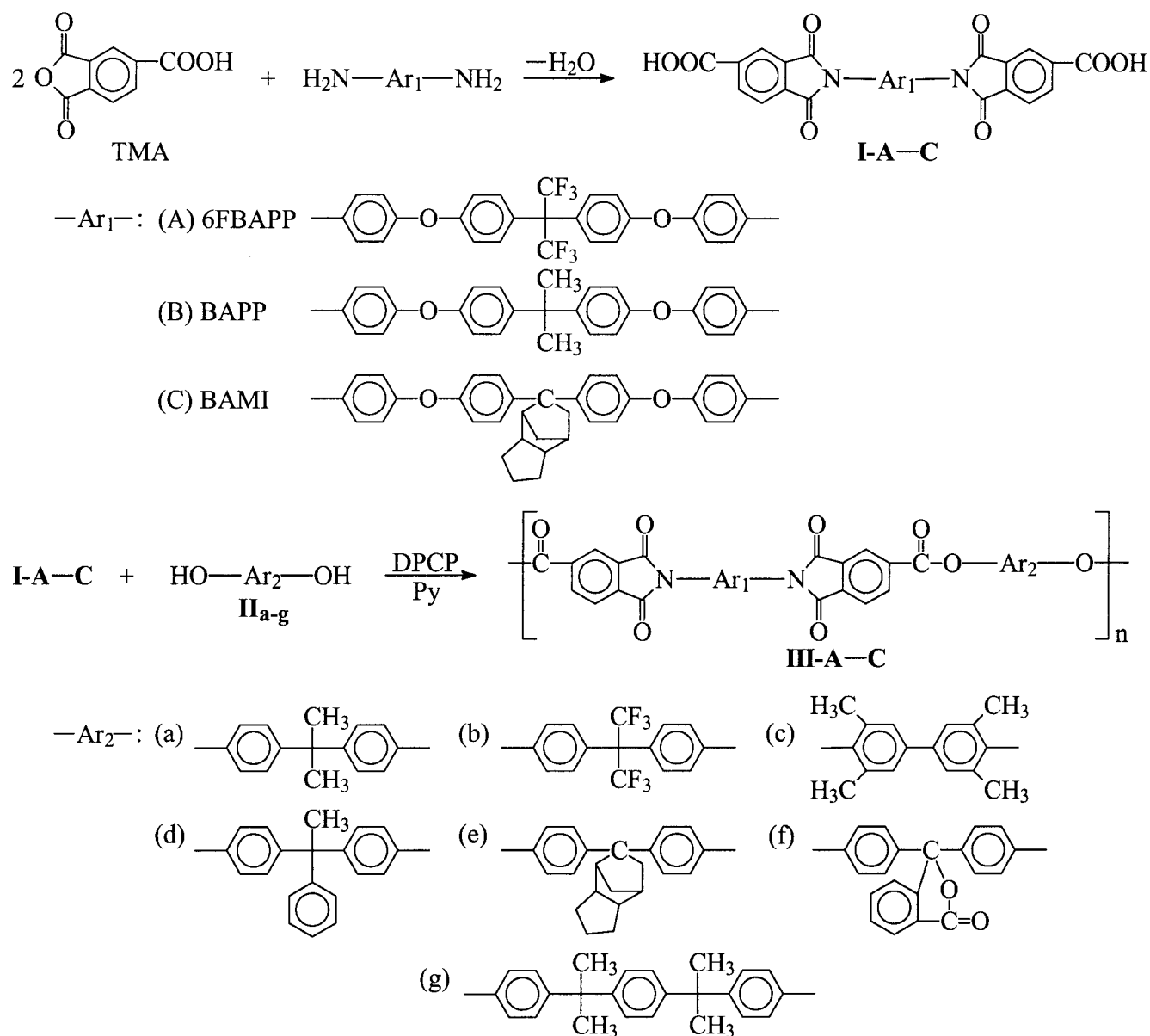
RESULTS AND DISCUSSION

Synthesis

The imide-containing dicarboxylic acids I-A–C were synthesized via a two-stage procedure that included ring-opening addition of the diamines (6FBAPP, BAPP, and BAMI) with two equivalent amounts of TMA, followed by cyclodehydration to the imidodicarboxylic acid by means of toluene–water azeotropic distillation, as shown in Scheme 1. The structures of diacids I-A–C were confirmed by elemental analysis

TABLE I
Synthesis and Elemental Analysis of Poly(arylate-imide)s

Polymer	Amount of Pyridine (mL)	η_{inh} (dL/g)	Polymer Appearance	Formula (M_w)	Elemental Analysis (%)			
					C	N	H	
III-A _a	6	0.51	String	$(C_{60}H_{36}O_{10}N_2F_6)_n$ (1058.94) _n	Calcd	68.05	2.65	3.43
					Found	67.85	2.65	3.38
III-A _b	6	0.41	String	$(C_{60}H_{30}O_{10}N_2F_{12})_n$ (1166.89) _n	Calcd	61.76	2.40	2.59
					Found	61.64	2.43	2.90
III-A _c	8	0.82	String	$(C_{61}H_{38}O_{10}N_2F_6)_n$ (1072.97) _n	Calcd	68.28	2.61	3.57
					Found	68.05	2.63	3.97
III-A _d	8	0.63	String	$(C_{65}H_{38}O_{10}N_2F_6)_n$ (1121.01) _n	Calcd	69.64	2.50	3.42
					Found	69.49	2.49	3.79
III-A _e	8	0.42	String	$(C_{67}H_{44}O_{10}N_2F_6)_n$ (1151.08) _n	Calcd	69.91	2.43	3.85
					Found	69.66	2.40	4.10
III-A _f	8	0.47	String	$(C_{65}H_{34}O_{12}N_2F_6)_n$ (1148.98) _n	Calcd	67.95	2.44	2.98
					Found	67.52	2.44	3.36
III-B _d	6	0.58	Lump	$(C_{65}H_{44}O_{10}N_2)_n$ (1013.07) _n	Calcd	77.06	2.77	4.38
					Found	76.20	2.47	4.40
III-B _e	6	0.54	String	$(C_{67}H_{50}O_{10}N_2)_n$ (1043.14) _n	Calcd	77.15	2.69	4.83
					Found	76.14	2.37	4.93
III-B _f	6	0.54	String	$(C_{65}H_{40}O_{12}N_2)_n$ (1041.04) _n	Calcd	74.99	2.69	3.87
					Found	74.25	2.38	3.83
III-C _a	6	0.44	String	$(C_{67}H_{50}O_{10}N_2)_n$ (1043.14) _n	Calcd	77.15	2.69	4.83
					Found	76.12	2.39	4.98
III-C _g	7	0.42	String	$(C_{76}H_{60}O_{10}N_2)_n$ (1161.32) _n	Calcd	78.60	2.41	5.21
					Found	77.03	2.45	5.74



Scheme 1 The synthesis of imide-containing dicarboxylic acids I-A-C, II_{a-g} and III-A-C.

and IR spectroscopy. The elemental analysis values were in good agreement with the calculated ones. The IR characteristic absorptions of I-A are shown in Figure 1. It exhibited the characteristic absorptions at 2500–3500 cm^{-1} , which is due to carboxylic acid $-\text{OH}$, and at 1782 and 1722 cm^{-1} , which are due to the asymmetrical and symmetrical $\text{C}=\text{O}$ stretching vibrations of the imide rings. Three series of novel poly(arylate-imide)s (III-A-C) were synthesized from diimide-diacids I-A-C and various bisphenols by means of direct polycondensation with DPCP as a condensing agent in the Py solution (Scheme 1). The synthesis conditions and results are summarized in Table I. All the reactions went smoothly in the homogeneous solution under the conditions listed in Table I.

The polymers were obtained in almost quantitative yield and were soluble in DMAc. Therefore, the measurement of the inherent viscosity was carried out without any difficulty, and the inherent viscosities of these polymers were in the range of 0.41–0.82 dL/g. All the poly(arylate-imide)s could be cast into transparent and tough films, indicating high molecular weight polymers. The formation of these poly(arylate-imide)s was characterized by their elemental analyses and IR and NMR spectroscopy. A typical IR spectrum is shown in Figure 1. The IR spectra of the polymers exhibited characteristic absorptions for the imide ring at 1782 and 1722 cm^{-1} , which is peculiar to the symmetrical and asymmetrical carbonyl stretching vibration. Bands of ester groups appeared at 1245 ($\text{C}-\text{O}-\text{C}$) and 1722 cm^{-1} ($\text{C}=\text{O}$), which overlapped

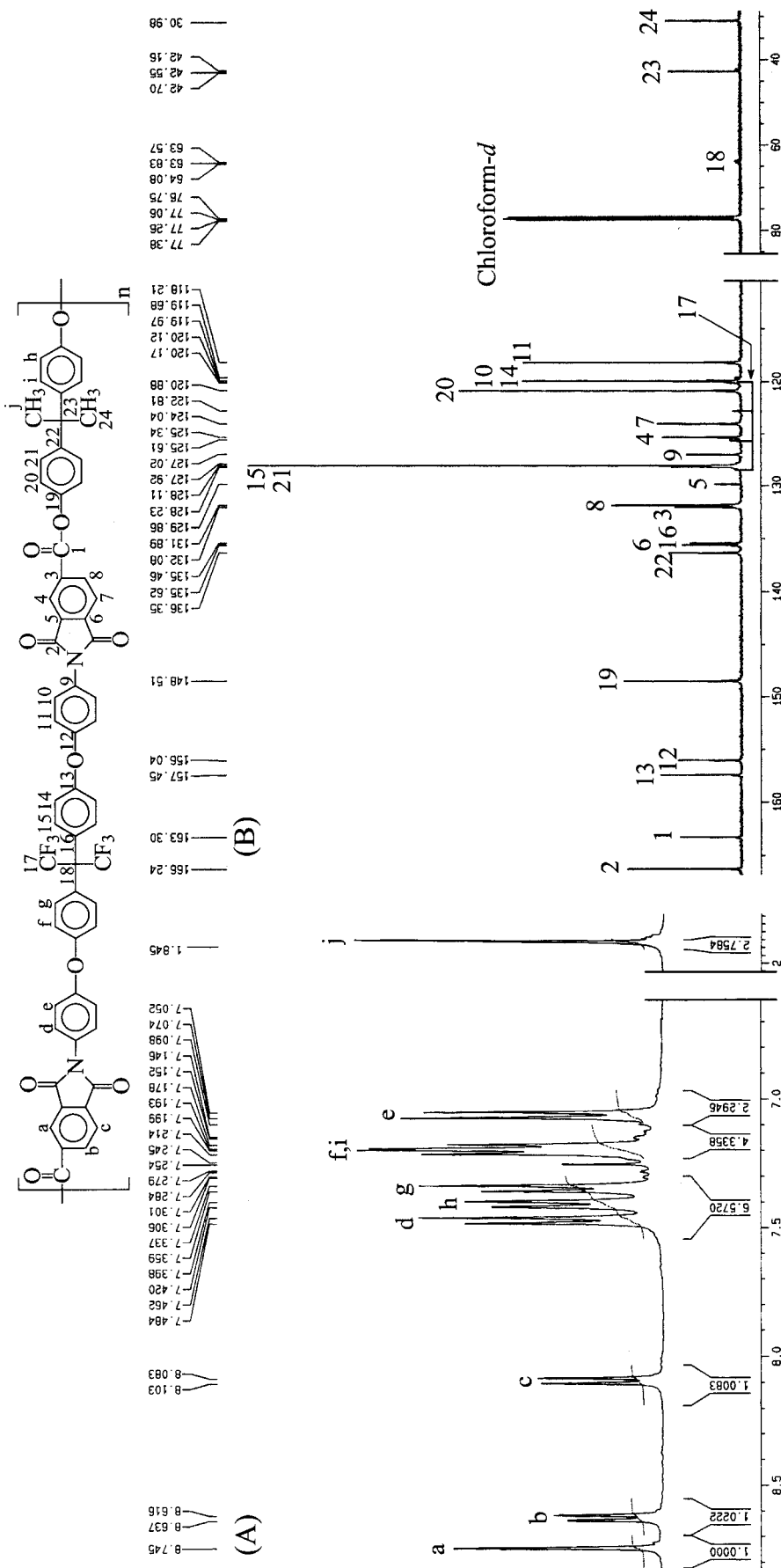


Figure 2 The (A) ¹H-NMR and (B) ¹³C-NMR spectra of poly(arylate-imide) III-A_a in CDCl₃.

TABLE II
Tensile Properties of Poly(arylate-imide) Films

Polymer	Strength at Break (MPa)	Elongation at Break (%)	Initial Modulus (GPa)
III-A _a	81	8	1.8
III-A _b	90	10	2.0
III-A _c	95	9	2.2
III-A _d	98	11	2.2
III-A _e	74	6	1.8
III-A _f	94	10	1.9
III-B _d	87	11	1.9
III-B _e	73	10	1.7
III-B _f	82	10	2.0
III-C _a	81	8	1.6
III-C _g	92	9	2.2

with one of the absorptions of the imide ring. The elemental analysis data of the poly(arylate-imide)s listed in Table I are generally in good agreement with the respective structures for hydrogen and nitrogen whereas the carbon analysis data were slightly lower than the theoretical ones, a phenomenon common to high-temperature polyimides.

In general, the structure identification of normal polyimides by the NMR spectra is difficult because of poor solubility. In this study the poly(arylate-imide)s had good solubility in low polarity solvents and could be identified by the NMR spectra. A typical NMR spectrum for III-A_a is shown in Figure 2. The protons H_a, H_b, and H_c adjacent to the ester group or imide ring resonated at the farthest downfield region, because of the inductive effect and resonance. The chemical shifts of H_e and H_f shown in the upfield shifting of their positions might be attributable to the electron-donating phenoxy groups in the ortho positions. The signal of H_j from the aliphatic protons is evident in the farthest upfield region. The ¹³C-NMR spectrum of III-A_a showed 28 signals, including 2 signals of the

carbonyl groups and 19 signals of the carbon of benzene. Furthermore, the splitting of the ¹³C signals caused by the couplings between carbon and fluorine were also observed in the spectrum. The magnitudes of the one-bond and two-bond carbon-fluorine couplings (¹J_{CF} and ²J_{CF}) are 280 and 25 Hz, respectively. All the spectroscopic data obtained were in good agreement with the expected structure.

Properties of polymers

The solubility of polymers III-A–C was tested qualitatively in various solvents at room temperature. These three series of polymers were easily soluble in halogenated hydrocarbons (CH₂Cl₂ and CHCl₃) and polar amide-type solvents, whose polarities were in the order of NMP > DMAc > DMF. In addition to highly polar solvents, they were also soluble in less polar solvents such as a protonic solvent (*m*-cresol), a basic solvent (Py), and even in dioxane and tetrahydrofuran at room temperature. The good solubility of these polymers might be due to the presence of the easily soluble groups of hexafluoroisopropyl, flexible isopropyl, or bulky polycyclic groups in the diimide-diacids I-A–C. The solubility behavior of the new poly(arylate-imide)s was compared to those of the corresponding poly(arylate-imide)s without pendent groups, and it also revealed that the new poly(arylate-imide)s had much better solubility than other poly(arylate-imide)s in many organic solvents. Thus, the solubility of the poly(arylate-imide)s was greatly improved by the introduction of bulky hexafluoroisopropyl, flexible isopropyl, and polycyclic groups into the polymer backbones.

All of the poly(arylate-imide)s could be cast into transparent, flexible, and tough films from polymer solutions of DMAc. The tensile properties of the polymer films were determined by an Instron machine, and the results are summarized in Table II. These films

TABLE III
Thermal Properties of Poly(arylate-imide)s

Polymer	TMA T _s (°C)	TGA			
		Decomposition Temperature (°C)		Char Yield (%)	Abnormal Range (°C)
		In Air	In Nitrogen		
III-A _a	214	508	518	43	531–598
III-A _b	209	517	522	46	543–584
III-A _c	248	481	500	46	512–552
III-A _d	226	505	511	47	523–595
III-A _e	240	496	497	35	496–596
III-A _f	244	501	503	42	524–578
III-B _d	145	506	506	49	518–614
III-B _e	159	470	492	36	506–599
III-B _f	200	485	498	47	508–595
III-C _a	199	479	483	41	507–621
III-C _g	198	454	450	51	470–595

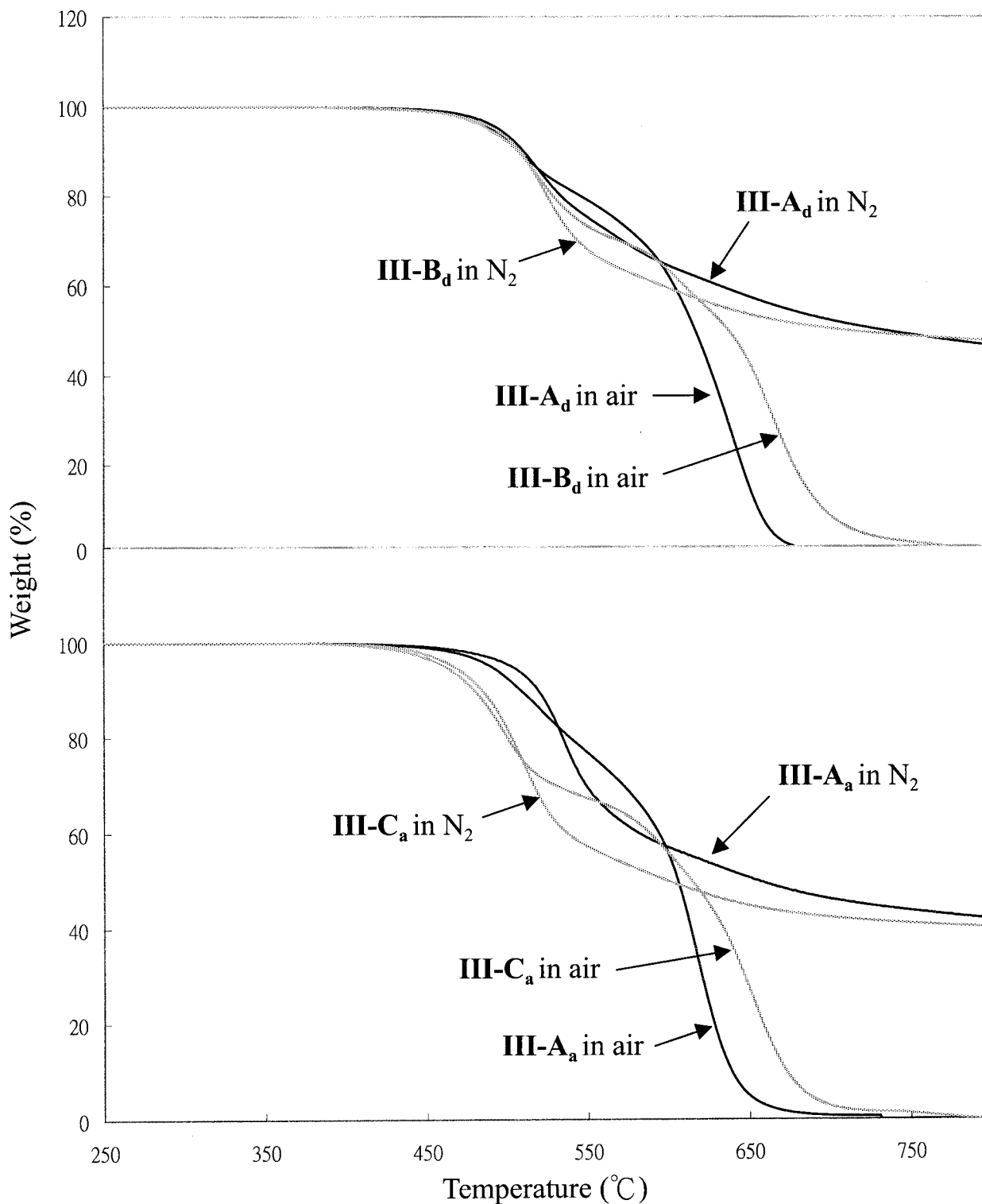


Figure 3 Typical TGA curves of poly(arylate-imide)s III-A_d, III-B_d, and III-C_a with a heating rate of 20°C/min.

had tensile strength at break of 73–98 MPa, tensile elongation at break of 6–11%, and initial moduli of 1.6–2.2 GPa. Taking the Ar₂ linkages into account, the introduction of aromatic pendant groups and phthalide groups into the molecular chains (III-A_{d,f} and

III-B_{d,f}) caused high tensile strength and greater elongation of the polymers. The three series of poly(arylate-imide)s all possessed good tensile properties, suggesting that these poly(arylate-imide)s could be applied as new materials for engineering plastics.

The thermal properties of all polymers were evaluated by TMA and TGA. The thermal behavior data of the poly(arylate-imide)s are shown in Table III. The softening temperatures (T_s) of the polymer film samples were measured with TMA by the penetration method. The T_s values were obtained from the onset temperature of the probe displacement on the TMA traces. The T_s values of these polymers were in the range of 145–248°C, depending on the structure of the bisphenol component and following a trend with increasing steric hindrance of the pendent groups. Polymer **III-A_c** containing tetramethyl-substituted biphenylene had the highest T_s value, and the bulky phthalide-group-containing polymers **III-A_f** and **III-B_f** also had higher T_s values than the other polymers. This phenomenon could be attributed to the restricted rotation about the bond joining the aromatic ring systems.

The thermal stability of the poly(arylate-imide) **III** series was studied by TGA. The temperatures at 10% weight loss (T_d) in nitrogen and air atmospheres were determined from original thermograms and are tabulated in Table III. The T_d values of these polymers were recorded in the range of 450–522°C in nitrogen and 454–517°C in air, which were reasonable values considering the aliphatic content of these polymers. The typical TGA curves of representative polymers in both air and nitrogen atmospheres are illustrated in Figure 3. It was interesting to note that in some cases the T_d in an air atmosphere was higher than that in nitrogen, and the decomposition rate was lower in air between 500 and 600°C. This might be attributable to an early weight-gained oxidation and possible oxidative interchain crosslinking of the aliphatic segments in the polymer chain when thermally degraded in air. The fluorine-containing poly(arylate-imide) **III-A_b** had a slightly better thermal stability than its non-fluoro analogue **III-A_a** because the C—F bond of the CF₃ group is stronger than the C—H bond of the CH₃ group.

For polymers in the same series, the T_d value of the poly(arylate-imide)s depended on the structure of the diamine component. Bulky pendent-group-containing **III-A_{d-f}** showed a thermostable order **III-A_d** > **III-A_f** > **III-A_e**, because the aromatic ring had better thermal stability than the aliphatic one; **III-A_d** also exhibited the highest char yield among them. Similar results could also be found among polymers **III-B_{d-f}**. Comparing the thermal stability of **III-A_{a-f}** with those of

III-B_{d-f} and **III-C_{a,g}**, the T_s and T_d values of most of the **III-A** series of poly(arylate-imide)s were much higher than **III-B_{d-f}** and **III-C_{a,g}**. Apparently, the great volume and high polarity of the 6F group overcame the effects of chain separation and disruption of hydrogen bonding caused by the bulky 6F group.

CONCLUSIONS

Three series of alternating aromatic poly(arylate-imide)s were successfully prepared by the direct polycondensation of three pendent-group-containing diimide-diacids with various bisphenols. These poly(arylate-imide)s had moderate to high molecular weights and showed higher thermal stability, higher T_s , good film-forming ability, excellent solubility in organic solvents, and outstanding tensile properties. Thus, they are considered to be promising, processable, and high-performing polymeric materials.

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