Synthesis and Characterization of New Poly(amide-imide)s Based on 1,4-Bis(trimellitimido)-2,5-dichlorobenzene

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ABSTRACT: A series of new aromatic poly(amide-imide)s were synthesized by the triphenyl phosphite-activated polycondensation of the diimide-diacid, 1,4-bis(trimellitimido)-2,5-dichlorobenzene (**I**), with various aromatic diamines in a medium consisting of *N*-methyl-2-pyrrolidone (NMP), pyridine, and calcium chloride. The poly(amide-imide)s had inherent viscosities of 0.88–1.27 dL g^{-1}. The diimide-diacid monomer (**I**) was prepared from 2,5-dichloro-*p*-phenylenediamine with trimellitic anhydride. All the resulting polymers were amorphous and were readily soluble in a variety of organic solvents, including NMP and *N,N*-dimethylacetamide. Transparent, flexible, and tough films of these polymers could be cast from *N,N*-dimethylacetamide or NMP solutions. Cast films had tensile strengths ranging from 92 to 127 MPa, elongations at break from 4 to 24%, and initial moduli from 2.59 to 3.65 GPa. The glass transition temperatures of these polymers were in the range of 256°–317°C, and the 10% weight loss temperatures were above 430°C in nitrogen. © 1999 John Wiley & Sons, Inc. J Appl Polym Sci 73: 271–278, 1999

Key words: aromatic poly(amide-imide)s; 1,4-bis(trimellitimido)-2,5-dichlorobenzene; solubility; thermal behavior

INTRODUCTION

The major limitations of aromatic polyimides for numerous applications are their high melting temperatures and limited solubility in organic solvents. To overcome these drawbacks, various copolyimides have been proposed. Poly(amide-imide)s (PAIs) represent one class of polymers that seems to have a good compromise between thermostability and processability.[1,2] Conventionally, trimellitic anhydride (TMA)-based PAIs are prepared in several ways, such as two-step polycondensation from the acid chloride of TMA, with aromatic diamines involving polyaddition and subsequent cyclodehydration,[3] low-temperature solution polycondensation of TMA-derived imide ring-preformed diacid chlorides and aromatic diamines,[4–7] and polycondensation of TMA or TMA-derived imide ring-containing dicarboxylic acids with diisocyanates.[8–10] Other synthetic routes of PAIs included the polymerization of *N,N′*-bis(trimethylsilyl)-substituted aromatic diamines with 4-chloroformylphthalic anhydride,[11] the palladium-catalyzed carbonylation and coupling of aromatic diamines and diimide-containing diiodides,[12] as well as the simultaneous catalytic amidation and imidization of TMA-derived *o*-ester acids.[13] Recently, we have successfully applied

phosphorylation polymerization technique[14] to the synthesis of high-molecular-weight PAIs by the direct polycondensation of imide-containing dicarboxylic acids with aromatic diamines using triphenyl phosphite and pyridine as condensing agents.[15–25] The direct polycondensation route avoids using moisture-sensitive acid chlorides or isocyanates, and provides significant advantages in laboratory preparation of aramids. Very recently, in a continuation of this study, we have been interested in the potential usefulness of substituent-containing *p*-phenylene structures as simultaneously bulky and symmetrical units in the polymer main chain. However, only a limited number of poly(ester-imide)s derived from diimide-diacid, 1,4-bis(trimellitimido)-2,5-dichlorobenzene, and bisphenols had been reported earlier[26] and have not been investigated in detail. The present article describes the synthesis of a series of novel PAIs from 1,4-bis(trimellitimido)-2,5-dichlorobenzene and various aromatic diamines. The solubility, tensile properties, crystallinity, and thermal properties of the obtained polymers will also be investigated.

To get more insight into the effect of the introduction of bulky group on properties of PAIs, the unsubstituted polymers based on 1,4-bis(trimellitimido)benzene was also synthesized in present study and the diimide-diacid was used to prepare PAIs to make a comparison of the solubility behavior with the previously described corresponding PAIs derived from 1,4-bis(trimellitimido)-2,5-dichlorobenzene.

EXPERIMENTAL

Materials

2,5-Dichloro-*p*-phenylenediamine (from TCI), TMA (from Wako), and triphenyl phosphite (from TCI) were used without further purification. Commercially obtained anhydrous calcium chloride was dried under vacuum at 150°C for 6 h. *N*-Methyl-2-pyrrolidone (NMP) and pyridine were purified by distillation under reduced pressure over calcium hydride and stored over 4 Å molecular sieve. *p*-Phenylenediamine (**IIa**; from Wako) and *m*-phenylenediamine (**IIb**; from TCI) were distilled under vacuum before use. All other diamines, such as 4,4′-oxydianiline (**IIc**; from TCI), 3,4′-oxydianiline (**IId**; from Teijin Ltd., Tokyo), 4,4′-methylenedianiline (**IIe**; from TCI), 4,4′-thiodianiline (**IIf**; from TCI), 1,4-bis(4-aminophen-

oxy)benzene (**IIg**; from TCI), and diamines {including 1,3-bis(4-aminophenoxy)benzene (**IIh**), 2,2-bis-[4-(4-aminophenoxy)phenyl]propane (**IIi**), and 2,2-bis[4-(4-aminophenoxy)phenyl]hexafluoropropane (**IIj**)}, were obtained from Chriskev Corp. and used without further purification.

Monomer Synthesis

1,4-Bis(trimellitimido)-2,5-dichlorobenzene (I)

A mixture of 5.31 g (30 mmol) of 2,5-dichloro-*p*-phenylenediamine and 11.55 g (60 mmol) of TMA were dissolved in 80 mL of dry NMP at 60°C and stirred for 1 h. About 30 mL of toluene were then added, and the mixture was heated with reflux for 3 h until ~ 1.1 mL of water was distilled off azeotropically under a Dean–Stark trap. Heating was continued to distill off the residue toluene. After cooling, the white precipitate of diimide-diacid (**I**) was isolated by filtration and washed with methanol. The product obtained was dried under vacuum affording 14.8 g (94% yield) of white powders; m.p. 448°–449°C [by differential scanning calorimetry (DSC)]. ANAL. Calcd for $C_{24}H_{10}N_2O_8Cl_2$: C, 54.86%; H, 1.90%; N, 5.33%. Found: C, 54.75%; H, 2.06%; N, 5.28%.

Polymerization

A typical example of polycondensation follows: a mixture of 0.108 g (1.0 mmol) of diamine **IIa**, 0.525 g (1.0 mmol) of diimide-diacid **I**, 0.44 g of calcium chloride, 6 mL of NMP, 1.2 mL of pyridine, and 0.7 mL of triphenyl phosphite was heated at 100°C for 3 h. The obtained polymer solution was trickled into 300 mL of methanol. The stringy polymer was washed thoroughly with methanol and hot water, collected by filtration, and dried at 100°C under vacuum. The yield was 0.59 g (99%). The inherent viscosity of the polymer was 1.23 dL g^{-1}, measured at a concentration of 0.5 g dL^{-1} in *N,N*-dimethylacetamide (DMAc) at 30°C.

Other PAIs were synthesized analogously.

Characterization

Infrared spectra were recorded on a Jasco FT/IR-7000 Fourier transform infrared spectrometer. Elemental analysis were run in a Perkin–Elmer model 240 C,H,N, analyzer. Inherent viscosities of all polymers were determined at a concentration of 0.5 g dL^{-1} in DMAc at 30°C using a Cannon–Fenske viscosimeter. Polymer films were

Scheme 1 Synthetic processes of poly(amide-imide)s.

prepared by slow evaporation of their DMAc solutions at 90°C for 12 h and then vacuum-dried at 150°C for 10 h. Thermogravimetric analysis (TGA) was conducted with a Rigaku thermal analysis station TAS-100. Measurements were performed with 10 ± 2 mg samples heated in flowing nitrogen (50 cm^3 min^{-1}) at a heating rate of 20°C min^{-1}. DSC analyses were performed on a Sinku Riko DSC-7000 differential scanning calorimeter coupled to a Sinku Riko TA-7000 thermal

Table I   Preparation of PAIs from Diimide-Diacids and Various Aromatic Diamines[a]

| | Amount of Reagents Used | | | | | |
Polymer	NMP (mL)	CaCl$_2$ (g)	Py (mL)	TPP (mL)	η_{inh}[b] (dL g^{-1})	Polymer Appearance
IIIa	6	0.44	1.2	0.7	1.23	Stringy
IIIb	5+1	0.4	1.2	0.7	0.95	Stringy
IIIc	5	0.35	1.2	0.6	1.02	Stringy
IIId	4+2	0.3	1.0	0.6	0.95	Stringy
IIIe	5	0.35	1.2	0.6	0.88	Stringy
IIIf	5+1	0.35	1.2	0.6	1.27	Stringy
IIIg	5+1	0.35	1.0	0.6	1.17	Stringy
IIIh	5+1	0.3	1.0	0.6	1.08	Stringy
IIIi	5+1	0.35	1.0	0.6	1.10	Stringy
IIIj	5	0.35	1.0	0.6	1.27	Stringy

Py, pyridine; TPP, triphenyl phosphite.
[a] Polymerization was conducted with 1.0 mmol of each monomer at 100°C for 3 h.
[b] Measured at a concentration of 0.5 g dL^{-1} in DMAc at 30°C.

analysis controller in flowing nitrogen (30 cm^3 min^{-1}) at a heating rate of 20°C min^{-1}. Wide-angle X-ray diffractograms were obtained on a Rigaku Geiger Flex D-Max IIIa, using Ni-filtered CuK$_\alpha$ radiation (40 kV, 15 mA), and the scanning rate was 4° min^{-1}. Measurements were performed with film specimens of $\sim$ 0.1 mm in thickness. An Instron Universal Tester Model 1130 with a load cell of 5 kg was used to study the stress-strain behavior of the samples at a drawing speed of 5 cm min^{-1}. Measurements were performed at room temperature ($\sim$ 20°C) with film specimens (0.5 cm wide, 6 cm long, and $\sim$ 0.1 mm thick), and an average of at least five individual determinations was used.

RESULTS AND DISCUSSION

Synthesis

1,4-Bis(trimellitimido)-2,5-dichlorobenzene (**I**), a novel PAI-forming diimide-diacid with preformed imide rings, was synthesized *via* the two-stage procedure that included ring-opening addition of 2,5-dichloro-*p*-phenylenediamine with two equivalent amounts of TMA, followed by cyclodehydration to the imidodicarboxylic acid by toluene-water azeotropic distillation (Scheme 1). The Fourier transform infrared spectrum (KBr) of the dicarboxylic acid (**I**) exhibited absorption bands at 3440 cm^{-1}–3206 cm^{-1} (acid —OH), 1779 cm^{-1} (imide, symmetric C=O stretching), and 1734

cm^{-1} (acid C=O stretching and asymmetric imide C=O stretching).

A series of new PAIs (**IIIa–j**) containing 2,5-dichloro-*p*-phenylene unit was prepared from diimide-diacid **I** and various aromatic diamines **IIa–j** by the direct polycondensation reaction using triphenyl phosphite and pyridine as condensing agents (Scheme 1). The reaction condition and results of the polycondensation are summarized in Table I. These polymers were obtained in almost quantitative yield with inherent viscosities of 0.88–1.27 dL g^{-1} measured at a concentration of 0.5 g dL^{-1} in DMAc at 30°C. The solubility of the polymer solution and the state of stirring affected the inherent viscosity of the resulting PAIs significantly. Generally, the molecular weight of the polymers obtained from the phosphorylation reaction is highly dependent on the reactant concentration. Higher molecular weights of these polymers could be obtained by using a higher initial reactant concentration and adding a proper amount of supplemental NMP into the viscous reaction medium before the formation of swollen gel. All the molecular weights of these polymers are sufficiently high to permit casting tough and flexible films.

The formation of PAIs was confirmed by elemental analysis and infrared spectroscopy. The elemental analysis values of these polymers are listed in Table II. In all cases, however, the found values of carbon were lower than the calculated values for the proposed structures. These may be attributed to

Table II Elemental Analysis of PAIs

Polymer	Formula (Molecular Weight)		Elemental Analysis[a] (%)			Moisture Intake (%)[b]
			C	H	N	
IIIa	$(C_{30}H_{14}N_4O_6Cl_2)n$	Calcd	60.33	2.37	9.38	4.33
	$(597.24)n$	Found	57.18	3.15	8.32	
		Corrected	59.66	3.01	8.68	
IIIb	$(C_{30}H_{14}N_4O_6Cl_2)n$	Calcd	60.33	2.37	9.38	4.49
	$(597.24)n$	Found	57.08	2.98	8.38	
		Corrected	59.88	2.84	8.79	
IIIc	$(C_{36}H_{18}N_4O_7Cl_2)n$	Calcd	62.71	2.63	8.13	3.42
	$(689.30)n$	Found	60.25	3.11	7.79	
		Corrected	62.31	3.00	8.06	
IIId	$(C_{36}H_{18}N_4O_7Cl_2)n$	Calcd	62.71	2.63	8.13	2.68
	$(689.30)n$	Found	60.40	3.06	7.46	
		Corrected	62.62	2.88	7.73	
IIIe	$(C_{37}H_{20}N_4O_6Cl_2)n$	Calcd	64.64	2.93	8.15	4.41
	$(687.31)n$	Found	61.79	3.26	7.52	
		Corrected	64.51	3.11	7.85	
IIIf	$(C_{36}H_{20}N_4O_6SCl_2)n$	Calcd	61.29	2.57	7.94	3.25
	$(705.37)n$	Found	59.30	3.00	7.84	
		Corrected	61.22	2.90	8.10	
IIIg	$(C_{42}H_{22}N_4O_8Cl_2)n$	Calcd	64.55	2.84	7.17	3.31
	$(781.36)n$	Found	62.41	3.20	7.33	
		Corrected	64.48	3.09	7.57	
IIIh	$(C_{42}H_{22}N_4O_8Cl_2)n$	Calcd	64.45	2.84	7.17	3.00
	$(781.36)n$	Found	62.61	3.27	7.42	
		Corrected	64.48	3.17	7.38	
IIIi	$(C_{51}H_{32}N_4O_8Cl_2)n$	Calcd	68.08	3.58	6.23	1.35
	$(899.45)n$	Found	67.16	3.89	5.97	
		Corrected	68.06	3.83	6.00	
IIIj	$(C_{51}H_{26}N_4O_8Cl_2F_6)n$	Calcd	60.79	2.60	5.56	1.28
	$(1007.45)n$	Found	60.01	2.99	5.25	
		Corrected	60.77	2.95	5.32	

[a] For C and N: corrected value = found value $\times$ (100% + moisture intake%). For H: corrected value = found value $\times$ (100% − moisture intake%).

[b] Moisture intake (%) = $(W - W_0)/W_0 \times 100\%$. W = weight of polymer sample after standing at room temperature. W_0 = weight of polymer sample after dried in vacuum at 100°C for 10 h.

the hygroscopic characteristics of the amide group. The moisture intake for PAIs **IIIa–j** was in the range of 1.28–4.49% at room temperature and normal pressure in our laboratory. The corrected values were in good agreement with the calculated ones after deducting the amount of moisture intake. The infrared spectra of the polymers exhibited characteristic absorptions for the imide ring at 1783 cm^{-1} and 1736 cm^{-1}, characteristic of the symmetrical and asymmetrical carbonyl stretching vibration. Bands of amide groups appeared at 3354 cm^{-1} (N—H) and 1671 cm^{-1} (C═O).

Properties of Polymers

Table III shows the qualitative solubility of the PAIs in various solvents. All the polymers were soluble in aprotic polar solvents such as DMAc, NMP, and *N,N*-dimethylformamide (DMF). Polymer **IIIa**, with rigid and symmetrical *p*-phenylene, also showed excellent solubility. This may be due to the amorphous nature of these polymers as evidenced by X-ray diffractograms discussed herein. All the polymers are insoluble in common organic solvents, such as chloroform, dichloromethane, acetone, and benzene; but, **IIIi** and **IIIj** are soluble in dioxane and tetrahydrofuran. The solubility behavior of the new PAIs derived from the diimide-diacid (**I**) and aromatic diamines were compared with those of the corresponding PAIs derived from 1,4-bis(trimellitimido)benzene without chloro-substituent summarized in Table IV. Thus, the solubility of PAIs was found to be

Table III   Solubility[a] of PAIs

Polymer	DMAc	DMAc + 5% LiCl	NMP	DMF	DMSO	Py	Dioxane	THF	CH$_2$Cl$_2$	Conc. H$_2$SO$_4$
IIIa	+	+	+	+	−	−	−	−	−	+
IIIb	+	+	+	+	+	±	−	−	−	+
IIIc	+	+	+	+	+	+	−	−	−	+
IIId	+	+	+	+	+	+	−	−	−	+
IIIe	+	+	+	+	+	+	−	−	−	+
IIIf	+	+	+	+	±	+	−	−	−	+
IIIg	+	+	+	+	+	+	−	−	−	+
IIIh	+	+	+	+	+	+	+	−	−	+
IIIi	+	+	+	+	+	+	+	+	−	+
IIIj	+	+	+	+	+	+	+	+	−	+

[a] Solubility: (+) soluble at room temperature; (−) insoluble; (±) partially soluble.
[b] DMF, *N,N*-dimethylformamide; DMSO, dimethylsulfoxide; Py, pyridine; THF, tetrahydrofuran.

greatly improved by the introduction of the bulky 2,5-dichloro-substituents into the *p*-phenylene unit along the polymer backbone.

The crystallinity of the prepared PAIs was measured by wide-angle X-ray diffraction studies. All the polymers showed completely amorphous patterns. Thus, the amorphous nature of these polymers was reflected in their excellent solubility, which is in agreement with the general rule that the solubility decreases with increasing crystallinity. In addition, by comparing the properties of the previously described polymer **IIIa** derived from 1,4-bis(trimellitimido)-2,5-dichlorobenzene and *p*-phenylenediamine with the PAI,[27] pre-

pared from 2,5-bis(trimellitimido)chlorobenzene and *p*-phenylenediamine, only the 2-chloro-*p*-phenylene-containing polymer showed semicrystalline behavior. Thus, the 2,5-dichloro-*p*-phenylene-containing PAIs had better solubility and higher glass transition temperatures (T_g) by ∼ 40°C than the 2-chloro-*p*-phenylene-containing polymers. This phenomenon is due to the fact that the former should be able to further lower the interchain interactions or prevent the polymer chains from close packing. Furthermore, by virtue of restricted rotation about the bond joining the aromatic ring systems, the 2,5-dichloro-*p*-phenylene-based polymers should have higher

Table IV   Solubility[a] of PAIs Derived from 1,4-Bis(trimellitimido)benzene

Polymer	DMAc	DMAc + 5% LiCl	NMP	DMF	DMSO	Py	Conc. H$_2$SO$_4$
Ia	−	±	−	−	−	−	+
Ib	−	+	−	−	−	−	+
Ic	−	+	−	−	−	−	+
Id	−	+	−	−	−	−	+
Ie	−	+	−	−	−	−	+
If	−	+	−	−	−	−	+
Ig	−	+	−	−	−	−	+
Ih	−	±	−	−	−	−	+
Ii	−	−	−	−	−	−	+
Ij	−	−	−	−	−	−	+

[a] Solubility: (+) soluble at room temperature; (−) insoluble; (±) partially soluble.
[b] DMF, *N,N*-dimethylformamide; DMSO, dimethylsulfoxide; Py, pyridine.

Table V   Tensile Properties of PAI Films[a]

Polymer	Yield Strength (MPa)	Tensile Strength (MPa)	Elongation at Break (%)	Initial Modulus (GPa)
IIIa	–	112	4	3.65
IIIb	–	117	5	3.13
IIIc	–	122	5	3.38
IIId	–	111	6	2.86
IIIe	–	120	5	3.26
IIIf	–	127	6	3.27
IIIg	–	113	6	3.01
IIIh	–	106	7	2.59
IIIi	116	113	15	2.98
IIIj	99	92	24	3.02

[a] Polymer films were prepared by slow evaporation of their DMAc solutions at 90°C for 12 h and vacuum-dried at 150°C for 10 h.

T_g's than the 2-chloro-p-phenylene-based analogs.

Transparent, flexible, and yellowish films could be cast from the DMAc solutions of all PAIs and were subjected to the tensile test. Tensile properties of these polymers are summarized in Table V. All specimens showed high yield or tensile strengths up to 127 MPa. Polymers **IIIi** and **IIIj** yield during tensile testing and had fairly high elongation to break, indicative of high toughness.

Thermal stability of the PAIs was studied by TGA. Typical TGA curves in both air (dotted line)

Figure 1   TGA and the second heating DSC curves for PAI **IIIj** with a heating rate of 20°C min^{-1}. (– – –) In air. (—) In nitrogen.

and nitrogen (solid line) atmospheres of representative PAI **IIIj** are shown in Figure 1. The temperatures of 10% weight loss in nitrogen and air atmosphere were determined from original thermograms and tabulated in Table VI. In general, all the polymers exhibited good thermal stability, with no significant weight loss up to temperatures of ~ 400°C in nitrogen; and their 10% weight loss temperatures were recorded in the range of 430°–525°C in nitrogen and 420°–505°C in air, respectively. The amount of carbonized residue (char yield) of PAIs in the nitrogen atmosphere was in the range of 53–58% weight, even at 800°C, showing high intrinsic fire-retardant characteristics.

Table VI   Thermal Behavior of PAIs

	DSC[a]	TGA Decomposition Temperature (°C)[b]		Wt % Residue[c] at 800°C in N$_2$
Polymer	T_g(°C)	In Air	In Nitrogen	
IIIa	[d]	460	475	55
IIIb	317	445	465	55
IIIc	308	440	450	56
IIId	298	410	445	56
IIIe	286	420	430	58
IIIf	289	440	480	58
IIIg	267	465	480	56
IIIh	256	450	445	56
IIIi	269	490	480	55
IIIj	273	505	525	53

[a] From the second heating of DSC measurements conducted with a heating rate of 20°C min^{-1} in nitrogen (N$_2$).
[b] Temperature at 10% weight loss was recorded by TGA at a heating rate of 20°C min^{-1}.
[c] Char yield was recorded by TGA at 800°C in a nitrogen atmosphere.
[d] No T_g was observed in DSC traces.

Table VI also summarizes the thermal transition data of the PAIs. Since the influence of residual water or solvent and history of thermal annealing are sometimes observed in the first heating scan of DSC, the samples were quenched from elevated temperatures to room temperature yield more amorphous samples so that the T_g's of these polymers could be easily measured in the second heating traces of DSC. The T_g's of the PAIs were in the range of 256°–317°C. A representative second heating DSC curve of polymer **IIIj** is shown in Figure 1.

CONCLUSIONS

A series of new high molecular weight aromatic PAIs, based on the diimide-diacid, 1,4-bis(trimellitimido)-2,5-dichlorobenzene, was successfully prepared by direct polycondensation with various aromatic diamines using triphenyl phosphite and pyridine as condensing agents. The PAIs were prepared noncrystalline. The introduction of the bulky and symmetrical 2,5-dichloro-*p*-phenylene unit into the polymer backbone lead to significantly improved solubility of the polymer in various organic solvents. Most of these polymers exhibited a desired combination of properties requiring high-performance materials that include excellent mechanical properties, high T_g's and thermal stability, and excellent solubility in organic solvents.

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