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Synthesis and characterization of new organo-soluble poly(amide-imide)s based on 1,4-bis(trimellitimido)-2,5-dimethylbenzene

Chin-Ping Yang[1,*], Guey-Sheng Liou[2], Chun-Chen Yang[1], Shin-Haur Chen[1]

[1] Department of Chemical Engineering, Tatung Institute of Technology,
40 Chungshan North Rd. 3rd Sec., Taipei, Taiwan, Republic of China
[2] Department of Chemical Engineering, I-Shou University, 1 Hsuen-Cheng Rd. 1st Sec.,
Ta-Hsu Hsiang, Kaohsiung 84008, Taiwan, Republic of China

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Summary

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-dimethylbenzene (**I**), with various aromatic diamines. The poly(amide-imide)s had inherent viscosities of 1.13-2.22 dL/g. Most of the resulting polymers showed an amorphous nature and were readily soluble in a variety of organic solvents. Transparent, flexible, and tough films of these polymers could be cast from DMAc or NMP solutions. Their cast films had tensile strengths ranging from 64 to 116 MPa, elongations at break from 6 to 20%, and initial moduli from 2.18 to 3.90 GPa. The glass transition temperatures of these polymers were in the range of 247-324°C.

Introduction

The development of heat resistant high performance polymers in the past decades has been quite dramatic and has drawn the attention of many polymer scientists all over the world. Wholly aromatic poly(amide-imide)s (PAIs) are an important class of materials having excellent resistant to high temperatures and favorable balance of other physical and chemical properties(1,2). However, these polymers are generally intractable and lack the properties essential for successful fabrication into useful forms due to their high melting or glass transition temperatures and limited solubility in organic solvents. Conventionally, trimellitic anhydride (TMA)-based PAIs could be 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), 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-chloroformlphthalic 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 reaction(14) to the synthesis of high-molecular-weight PAIs by the direct polycondensation of imide-containing dicarboxylic acids with aromatic diamines using triphenyl phosphite (TPP) and pyridine (Py) as condensing agents(15-19). 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 are interested in the potential usefulness of substituent-containing *p*-phenylene structure as simultaneously bulky and symmetrical unit in the polymer main chain. However, only a limited number of poly(ester-imide)s derived from the diimide-diacid, 1,4-bis(trimellitimido)-2,5-dimethylbenzene, and bisphenols had been reported earlier(20) and have not been investigated in detail.

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[*] Corresponding author

The present article describes the synthesis of a series of novel poly(amide-imide)s from TMA, 2,5-dimethyl-*p*-phenylenediamine, and other aromatic diamines. The solubility, tensile properties, crystallinity, and thermal properties of the obtained polymers will also be investigated. In order to get more insight into the effect of the introduction of bulky group on properties of poly(amide-imide)s, poly(amide-imide)s derived from 1,4-bis(trimellitimido)benzene(21) was also discussed to make a comparison with above mentioned corresponding poly(amide-imide)s derived from 1,4-bis(trimellitimido)-2,5-dimethylbenzene.

Experimental

Materials

2,5-Dimethyl-*p*-phenylenediamine (from TCI), trimellitic anhydride (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. *p*-Phenylenediamine (**IIa**; from Wako) and *m*-phenylenediamine (**IIb**, from TCI) were distillated under vacuum before use. All other reagent-grade diamines were used without further purification.

Monomer Synthesis

1,4-Bis(trimellitimido)-2,5-dimethylbenzene (**I**)

A mixture of 4.09g (30 mmol) of 2,5-dimethyl-*p*-phenylenediamine and 11.55g (60 mmol) of trimellitic anhydride were dissolved in 80 mL of dry NMP at 60°C and stirred for 1 h. About 30 mL of toluene was then added, and the mixture was heated with reflux for 3 h until about 1.1 mL of water was distillated 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 13.10g (90% yield) of white powders; mp 440-441°C (by DSC).

ANAL. Calcd for $\mathbf{C_{26}H_{16}N_2O_8}$: C, 64.47%; H, 3.33%; N, 5.78%. Found: C, 64.44%; H, 3.56%; N, 5.53%.

Polymerization

A typical example of polycondensation follows: A mixture of 0.112g (1.0 mmol) of diamine **IIc**, 0.484g (1.0 mmol) of diimide-diacid **I**, 0.30g of calcium chloride, 4.5 mL of NMP, 1.0 mL, of pyridine, and 0.6 mL of triphenyl phosphite was heated at 100°C for 3 h. The obtained polymer solution was trickled into 200 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 quantitative. The inherent viscosity of the polymer was 1.13 dL/g, measured at a concentration of 0.5 g/dL in DMAc at 30°C.

Other Poly(amide-imide)s were synthesized analogously.

Characterization

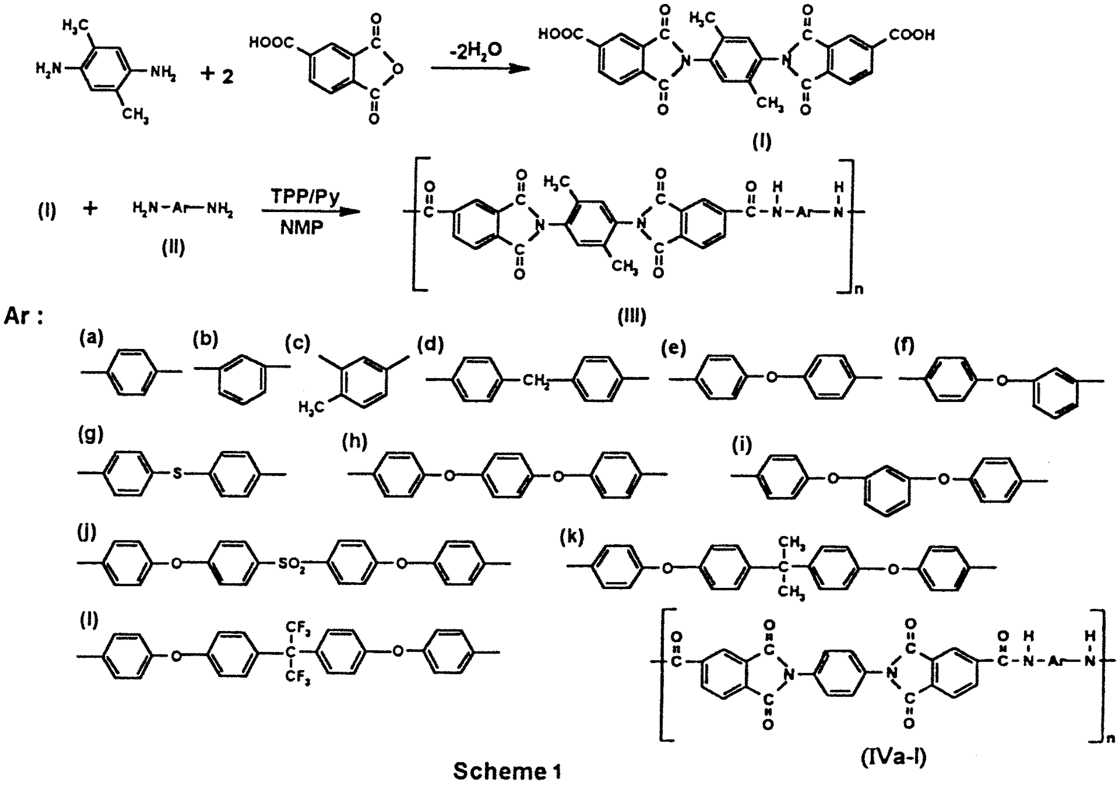
Infrared spectra were recorded on a Jasco FT/IR-7000 Fourier transform infrared spectrometer. Elemental analysis were run in a Perkin-Elmer model 240 C,H,N, analyzer. Inherent viscosities of all polymers were determined at 0.5 g/dL concentration using Cannon-Fenske viscosimeter. Thermogravimetric analysis 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) at a heating rate of 20°C/min.

Differential scanning calorimetry (DSC) analyses were performed on a Sinku Riko DSC-7000 differential scanning calorimeter coupled to a Sinku Riko TA-7000 thermal analysis controller in flowing nitrogen (30 cm^3/min) at a heating rate of 20°C/min. Wide-angle X-ray diffractograms were obtained on a Rigaku Geiger Flex D-Max IIIa, using Ni-filtered CuKα radiation (40 kV, 15 mA), and the scanning rate was 4°/min. Measurements were performed with film specimens of about 0.1 mm in thickness. An Instron Universal Tester Model 1130 with a load cell 5 kg was used to study the stress-strain behavior of the samples at a drawing speed of 5 cm/min were used for this study. Measurements were performed at room temperature (ca. 20°C) with film specimens (0.5 cm wide, 6 cm long, and about 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-dimethylbenzene (**I**), the novel poly(amide-imide)s-forming diimide-diacid with preformed imide rings, was synthesized via the two-stage procedure that included ring-opening addition of 2,5-dimethyl-p-phenylenediamine with two equivalent amount of trimellitic anhydride, followed by cyclodehydration to the imidodicarboxylic acid by toluene-water azeotropic distillation (Scheme 1). The FT-IR spectrum (KBr) of the dicarboxylic acid (**I**) showed absorption bands at 3436 cm^{-1} (acid -OH), 1700 cm^{-1} (acid C=O stretching), 1777 cm^{-1} (imide, symmetric C=O stretching), 1729 cm^{-1} (asymmetric imide C=O stretching).



Scheme 1

A series of new poly(amide-imide)s (**IIIa-1**) containing 2,5-dimethyl-*p*-phenylene unit were prepared from diimide-diacid **I** and various aromatic diamines **IIa-1** 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 1. These polymers were obtained in almost quantitative yield with inherent viscosities of 1.13-2.22 dL/g for **IIIa-1** and 0.19-0.51 dL/g for **IVb-k**. The solubility of the polymer solution and the state of stirring affected the inherent viscosity of the resulting poly(amide-imide)s significantly. Generally, the molecular weight of the polymers obtained from the phosphorylation reaction is highly dependent on the reactant concentration. Higher molecular weights of polymers **III** 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 polymers **III** are sufficiently high to permit casting tough and flexible films, but unsubstituted polymers **IV** produced a powdery precipitate during polymerization.

Table 1. Preparation of Poly(amide-imide)s from Diimide-diacid and Various Aromatic Diamines[a]

Polymer	Amount monomer (m mole)	Amount of Reagents Used NMP (ml)	Additional NMP(ml)	$CaCl_2$ (g)	Py (ml)	TPP (ml)	η_{inh}[b] (dL/g)	η_{inh}(**IVb-k**) (dL/g)
IIIa	1.0	6.0	3.5	0.45	1.0	0.6	2.22[c]	
IIIb	1.9	9.5	4.0	0.60	2.0	1.2	1.23	(0.25[d])
IIIc	1.0	4.5	0	0.30	1.0	0.6	1.13	(0.66[c])
IIId	1.5	7.0	1.0	0.45	1.5	0.9	1.23	(0.32[c])
IIIe	1.5	7.5	4.0	0.45	1.5	0.9	1.53	(0.47[c])
IIIf	1.5	7.5	0	0.45	1.5	0.9	1.15	(0.33[c])
IIIg	1.5	7.0	0	0.45	1.5	0.9	1.23	(0.31[d])
IIIh	1.5	7.5	2.0	0.45	1.5	0.9	1.54	(0.25[d])
IIIi	1.5	7.5	5.0	0.45	1.5	0.9	1.90	(0.19[d])
IIIj	1.5	7.0	5.0	0.45	1.5	0.9	1.55	(0.51[c])
IIIk	1.5	7.5	5.0	0.45	1.5	0.9	1.80	(0.43[c])
IIIl	1.5	7.5	0	0.45	1.5	0.9	1.20	

a) Polymerization was carried out at 100℃ for 3h.
b) Measured at a concentration of 0.5 g/dL in DMAc at 30℃.
c) Measured at a concentration of 0.5 g/dL in DMAc+5%LiCl at 30℃.
d) Measured at a concentration of 0.5 g/dL in conc. H_2SO_4 at 30℃.

The formation of poly(amide-imide)s was confirmed by elemental analysis and IR spectroscopy. The elemental analysis values of these polymers were in good agreement with the calculated ones after deducting the amount of moisture intake (Table 2). The IR spectra of the polymers exhibited characteristic absorptions for the imide ring at 1781 and 1725 cm^{-1}, peculiar to the symmetrical and asymmetrical carbonyl stretching vibration. Bands of amide groups appeared at 3352 cm^{-1} (N-H) and 1665 cm^{-1} (C=O).

Table 2. Elemental analysis of poly(amide-imide)s

Polymer	Formula (M. W.)		Elemental Analysis[a](%) C	H	N	Moisture Intake[b](%)
IIIa	$(C_{32}H_{20}N_4O_6)_n$	Calcd	69.06	3.62	10.07	
	$(556.53)_n$	Found	65.47	3.82	9.55	5.2
		Corrected	68.87	3.62	10.05	
IIIb	$(C_{32}H_{20}N_4O_6)_n$	Calcd	69.06	3.62	10.07	
	$(556.53)_n$	Found	65.40	3.81	9.57	5.3
		Corrected	68.87	3.61	10.08	
IIIc	$(C_{33}H_{22}N_4O_6)_n$	Calcd	69.47	3.89	9.82	
	$(570.56)_n$	Found	65.85	4.09	9.31	5.2
		Corrected	69.27	3.88	9.79	
IIId	$(C_{39}H_{26}N_4O_6)_n$	Calcd	72.44	4.05	8.66	
	$(646.66)_n$	Found	69.26	4.23	8.28	4.4
		Corrected	72.31	4.04	8.64	
IIIe	$(C_{38}H_{24}N_4O_7)_n$	Calcd	70.37	3.73	8.64	
	$(648.63)_n$	Found	67.20	3.90	8.25	4.5
		Corrected	70.22	3.72	8.62	
IIIf	$(C_{38}H_{24}N_4O_7)_n$	Calcd	70.37	3.73	8.64	
	$(648.63)_n$	Found	67.34	3.90	8.27	4.3
		Corrected	70.24	3.73	8.63	
IIIg	$(C_{38}H_{24}N_4O_6S)_n$	Calcd	68.67	3.64	8.43	
	$(664.69)_n$	Found	66.13	3.77	8.12	3.7
		Corrected	68.58	3.63	8.42	

(Table 2. continued)

IIIh	$(C_{44}H_{28}N_4O_8)_n$	Calcd	71.35	3.81	7.56	
	$(740.73)_n$	Found	68.78	3.95	7.27	3.6
		Corrected	71.26	3.81	7.53	
IIIi	$(C_{44}H_{28}N_4O_8)_n$	Calcd	71.35	3.81	7.56	
	$(740.73)_n$	Found	69.36	3.92	7.35	2.8
		Corrected	71.30	3.81	7.56	
IIIj	$(C_{50}H_{32}N_4O_{10}S)_n$	Calcd	68.18	3.66	6.36	
	$(880.88)_n$	Found	66.07	3.77	6.16	3.1
		Corrected	68.12	3.65	6.35	
IIIk	$(C_{53}H_{38}N_4O_8)_n$	Calcd	74.12	4.46	6.52	
	$(858.91)_n$	Found	71.63	4.70	5.87	3.3
		Corrected	74.07	4.54	6.07	
IIIl	$(C_{53}H_{32}N_4O_8F_6)_n$	Calcd	65.84	3.34	5.79	
	$(966.85)_n$	Found	64.02	3.67	5.30	2.8
		Corrected	65.79	3.57	5.95	

a) For C and N: Corrected value = found value × (100% + moisture intake %). For H: Corrected value = found value × (100% – moisture intake %).

b) Moisture intake (%) = $[(W-W_0)/W_0]\times 100\%$; W = weight of polymer sample after standing at room temperature for 3 days, and W_0 = weight of polymer sample after dried in vacuum at 100℃ for 10 h.

Properties of Polymers

Table 3 shows the qualitative solubility of the PAIs in various solvents. Most polymers were soluble in aprotic polar solvents such as DMAc, NMP, DMF, and even in less polar solvents like *m*-cresol and pyridine. Polymer **IIIa** with rigid and symmetrical *p*-phenylene unit had somewhat limited solubility. This may be due to the semi-crystalline nature of these polymers as evidenced by X-ray diffractograms. All the polymers are insoluble in common organic solvents such as chloroform, acetone, and benzene. The solubility behavior of the new poly(amide-imide)s derived from the diimide-diacid (**I**) and aromatic diamines were compared with those of the corresponding poly(amide-imide)s derived from 1,4 bis(trimellitimido)benzene without methyl-substituent also summarized in Table 3. Thus, the solubility of poly(amide-imide)s was found to be greatly improved by the introduction of bulky 2,5-dimethyl-substituent into phenylene unit along polymer backbone.

Table 3. Solubility of Poly(amide-imide)s

Polymer	DMAc	DMAc +5%LiCl	NMP	DMF	DMSO	*m*-cresol	Pyridine
IIIa	–	+	–	–	–	–	–
IIIb~f	+	+	+	+	+	+	+
IIIg	+	+	+	–	–	+	–
IIIh,i	+	+	+	+	+	–	–
IIIj~l	+	+	+	+	+	+	+
IVb	–	–	–	–	–	–	–
IVc~g	–	+h	–	–	–	–	–
IVh,i	–	–	–	–	–	–	–
IVj,k	–	+h	–	–	–	–	–

Solubility : (+) soluble at room temperature ; (+h) soluble on heating ; (–) insoluble. DMAc : *N,N*-dimethylacetamide ; DMF : *N,N*-dimethylformamide ; NMP : *N*-methyl-2-pyrrolidone ; DMSO : dimethyl sulfoxide.

The crystallinity of the prepared poly(amide-imide)s was measured by wide-angle X-ray diffraction scans. Typical diffraction patterns for some representative polymers are presented in Figure 1. Polymer **IIIa** showed a fair degree of crystallinity. This call be attributed to the existence of rigid planar imide segments in the polymer backbones that led to a better packing of polymer chain, whereas all of the other polymers showed completely amorphous pattern. Thus, the amorphous nature of these polymers was reflected

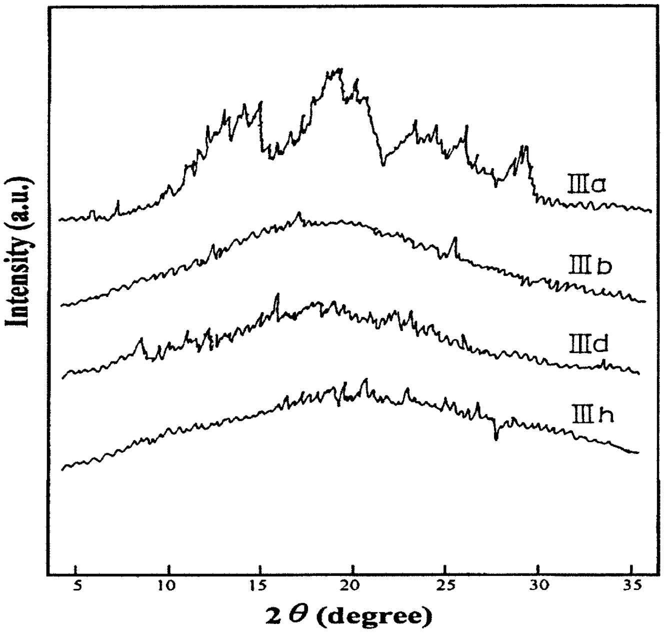


Figure. 1: Wide-angle X-ray diffraction patterns of poly(amid-imide)s.

in their excellent solubility, which is in agreement with the general rule that the solubility decreases with increasing crystallinity. In addition, to make a comparison of the solubility of the above mentioned polymer **III** derived from 1,4-bis(trimellitimido)-2,5-dimethylbenzene with the corresponding poly(amide-imide)s **IV**, it was found that the 2,5-dimethyl-*p*-phenylene-containing poly(amide-imide)s had better solubility than the *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.

Transparent, flexible, and yellowish films could be cast from the DMAc or NMP solutions of most PAIs and were subjected to tensile test. The tensile properties of these PAIs are summarized in Table 4. These films had tensile strength of 64-116 MPa, elongation to break of 6-20%, and initial modulus of 2.18-3.90 GPa. Some of polymers yielded during tensile testing and had fairly high elongation to break, indicative of high toughness.

The thermal stability of the poly(amide-imide)s was studied by thermogravimetric analysis (TGA). Typical TGA curves in both air (dotted line) and nitrogen (solid line) atmospheres of representative poly(amide-imide)s **IIIg** are shown in Figure 2. The temperatures of 10% weight loss in nitrogen and air atmosphere were determined from

Table 4. Tensile Properties of Poly(amide-imide) Films[a]

Polymer	Yield Strength (MPa)	Tensile Strength (MPa)	Elengation at Break (%)	Initial Modulus (GPa)
IIIb	–	109	6	3.90
IIIc	–	116	8	3.10
IIId	112	104	12	2.70
IIIe	106	101	13	2.66
IIIf	109	100	20	2.80
IIIg	–	83	6	2.40
IIIh	91	88	15	2.47
IIIi	–	64	12	2.18
IIIj	–	81	7	2.39
IIIk	95	92	8	2.39
IIIl	–	68	6	2.23

a) Films were prepared by slow evaporation of their polymer solutions of DMAc at 90°C for 12h, followed by vacuum dried at 150°C for 10h.

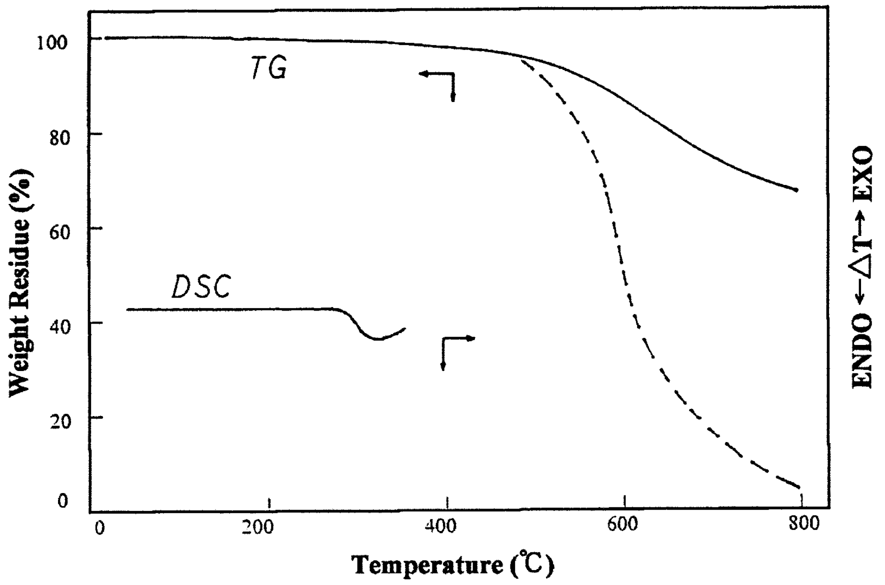


Figure. 2: TGA and the second heating DSC curves for poly(amide-imide) IIIg with a heating rate of 20℃/min; (...) in air, (—) in nitrogen.

Table 5. Thermal Behavior of Poly(amide-imide)s

		TGA		
	DSC[a]	Decomposition Temperature (℃)[b]		Wt % Residue[c]
Polymer	T_g(℃)	In air	In nitrogen	at 800℃
IIIa (IVa)	–[d]	500	535	52
IIIb (IVb)	317 (–)	490 (498)	520 (488)	51 (50)
IIIc (IVc)	324 (303)	470 (527)	505 (519)	58 (63)
IIId (IVd)	301 (–)	475 (500)	535 (489)	56 (51)
IIIe (IVe)	277 (–)	470 (491)	540 (482)	56 (48)
IIIf (IVf)	278 (–)	470 (499)	520 (484)	59 (49)
IIIg (IVg)	294 (–)	495 (516)	530 (542)	62 (57)
IIIh (IVh)	306 (–)	470 (508)	515 (538)	59 (53)
IIIi (IIIi)	260 (–)	495 (508)	535 (535)	59 (54)
IIIj (IVj)	268 (–)	485 (517)	520 (518)	51 (55)
IIIk (IVk)	247 (286)	475 (505)	515 (500)	56 (48)
IIIl (IVl)	254	520	540	57

a) From the second heating of DSC measurements conducted with a heating rate of 20℃/min in nitrogen.

b) Temperature at 10% weight loss was recorded by TGA at a heating rate 20℃/min in nitrogen.

c) Char yield was recorded by TGA at 800℃ under nitrogen atmosphere.

d) No T_g was observed in DSC traces.

original thermograms and tabulated in Table 5. In general, all the polymers exhibited good thermal stability with no significant weight loss up to temperatures of approximately 400°C in nitrogen, and their 10% weight loss temperatures (T_ds) were recorded in the range of 505-540°C in nitrogen and 470-520°C in air, respectively. The amount of carbonized residue (char yield) of poly(amide-imide)s in nitrogen atmosphere was in the range of 51-62% weight even at 800°C, showing high intrinsic fire-retardant characteristics. Because the methyl groups on the benzene were easy decomposed, polymers **IV** had higher T_ds than polymers **III**. However, T_ds in nitrogen and char yields of some polymers **III** were found to be higher than corresponding polymers **IV**, indicative of outstanding thermal stability of polymers **III**.

Table 5 also summarizes the thermal transition data of the poly(amide-imide)s. Since the influence of residual water or solvent and history of thermal annealing are sometimes observed in the first heating scan of DSC. Quenching from the elevated temperatures to room temperature yields more amorphous samples so that in most cases the glass transition temperatures (T_gs) could be easily measured in the second heating traces of DSC. The T_gs of the poly(amide-imide)s **III** were in the range of 247-324°C, but most of poly(amide-imide)s **IV** showed no T_g below 400°C. This difference may be due to the substituted methyl group along the phenyl moiety. Representative second heating DSC curve of polyer **IIIg** is shown in Figure 2.

Conclusion

A series of new high molecular weight aromatic poly(amide-imide)s based on the diimide-diacid, 1,4-bis(trimellitimido)-2,5-dimethylbenzene, were successfully prepared by direct polycondensation with various aromatic. Typical poly(amide-imide)s were noncrystalline. The introduction of bulky and symmetrical 2,5-dimethyl-*p*-phenylene unit into the polymer backbone lead to significantly improve solubility of the polymer in various organic solvents. Most of these polymers exhibited a desired combination of properties requiring for high performance materials that include excellent mechanical properties, high Tgs and thermal stability, excellent solubility in organic solvents.

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