New Poly(amide-imide) Syntheses. V. Preparation and Properties of Poly(amide-imide)s Based on the Diimide–diacid Condensed from 2,2-Bis[4-(4-aminophenoxy)phenyl]propane and Trimellitic Anhydride

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SYNOPSIS

A dicarboxylic acid (I) bearing two pre-formed imide rings, was prepared from the condensation of 2,2-bis[4-(4-aminophenoxy)phenyl]propane and trimellitic anhydride. A new family of poly(amide-imide)s having inherent viscosities of 0.53–1.68 dL/g was prepared by the triphenyl phosphite activated polycondensation from the diimide–diacid I with various aromatic diamines in a medium consisting of N-methyl-2-pyrolidone (NMP), pyridine, and calcium chloride. Most of the resulting polymers showed an amorphous nature and were readily soluble in polar solvents such as NMP and N,N-dimethylacetamide. All the soluble poly(amide-imide)s afforded transparent, flexible, and tough films. The glass transition temperatures of these poly(amide-imide)s were in the range of 237–293°C and the 10% weight loss temperatures were above 508°C in nitrogen. © 1993 John Wiley & Sons, Inc.
Keywords: poly(amide-imide)s • 2,2-bis[4-(4-aminophenoxy)phenyl]propane • direct polycondensation • triphenyl phosphite

INTRODUCTION

Because of their remarkable thermal stability and other excellent physical properties, linear aromatic polyimides are well recognized as a class of engineering materials. However, poor handling and intractable characteristics have been major problems due to high melting point and insolubility. To extend the utility of such high-performance materials, it has been a long-desired goal to synthesize soluble polymers without appreciable loss in their thermomechanical properties. It is known that the synthesis of alternative poly(amide-imide)s can offer a compromise between excellent thermal stability and processability.

Various approaches have been carried out successfully in the synthesis of poly(amide-imide)s.[1-8] A recently developed direct polycondensation method reported by Yamazaki et al.[9] using triphenyl phosphite as a condensation agent has been used to prepare high molecular weight poly(amide-imide)s from preformed imide-containing dicarboxylic acids and aromatic diamines.[10-15] In earlier articles,[16-19] we successfully applied this technique to the synthesis of a new series of poly(amide-imide)s containing tetraphenylthiophene or cardo linkage in the polymer backbone to make the polymer more amenable to its processing. In the present article, the diimide–diacid, 2,2-bis[4-(4-trimellitimidophenoxy)phenyl]propane (I), was first prepared by the condensation of 2,2-bis[4-(4-aminophenoxy)phenyl]propane (diamine II_n) and trimellitic anhydride, which was then directly polycondensed with various aromatic diamines by triphenyl phosphite forming a series of novel poly(amide-imide)s. The isopropylidene [$C(CH_3)_2$] and arylene ether-containing diamine II_n could be easily prepared by condensation of bisphenol A [2,2-bis(4-hydroxyphenyl)propane] and p-chloronitrobenzene followed by reduction of the dinitro compound. Introduction of an alkylene linkage in the aromatic polymer backbone is expected to improve

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fracture toughness by dissipating the impact energy due to mobility flexure, although this causes some sacrifice in thermooxidative stability. The solubility, tensile properties, thermal properties, and crystallinity of the resultant novel poly(amide-imide)s will be discussed.

EXPERIMENTAL

Materials

Trimellitic anhydride was used without previous purification. p-Phenylenediamine (II_a) and m-phenylenediamine (II_b) were vacuum-distilled before use. According to the reported method,[20] 2,5-bis(4-aminophenyl)-3,4-diphenylthiophene (II_o) was prepared starting from tetraphenylthiophene, which was easily obtained by the reaction of benzyl chloride with powdered sulfur at an elevated temperature. All other diamines II_{c-m} were of high purity when received from the manufacturers and used without any further purification. Commercially obtained calcium chloride was dried under vacuum at 180°C for 10 h. N-Methyl-2-pyrrolidone (NMP), N,N-dimethylformamide (DMF), and pyridine were purified by distillation under reduced pressure over calcium hydride and stored over 4 Å molecular seives. Triphenyl phosphite was purified by vacuum-distillation.

Synthesis of 2,2-Bis[4-(4-aminophenoxy)phenyl]propane (II_n)

A mixture of 104 g (0.5 mol) of bisphenol A, 185 g (1.175 mol) of p-chloronitrobenzene, 190 g (1.375 mol) of anhydrous K_2CO_3, and 500 mL of dry DMF was refluxed at 160°C on an oil bath for 8 h and cooled. The mixture was then poured into 500 mL of 1 : 1 H_2O–C_2H_5OH. The precipitate was collected by filtration and recrystallized from glacial acetic acid to afford 195 g (83%) of yellow crystals. One hundred grams of the dinitro compound, 0.6 g of 10% Pd — C, and 850 mL of ethanol were introduced into a three-necked flask to which 220 mL of hydrazine monohydrate was added dropwise over a period of 1 h at 85°C. After the addition was complete, the reaction was continued to reflux at 85°C for another 6 h. The mixture was then filtered to remove Pd — C and cooled to afford 68.8 g (79%) of colorless needles; mp 123–124°C (lit.[21] mp 126°C). The infrared (IR) spectrum (KBr): 3400, 3340 cm^{-1} (NH_2); 1220 cm^{-1} (C — O — C).

Synthesis of 2,2-Bis[4-(4-trimellitimidophenoxy)phenyl]propane (I)

A mixture of 41.1 g (0.1 mol) of 2,2-bis[4-(4-aminophenoxy)phenyl]propane (II_n) and 38.4 g (0.2 mol) of trimellitic anhydride were heated in 250 mL of dry DMF to 60°C for 1 h. About 100 mL of toluene was then added, and the mixture was heated at reflux for about 3 h until about 3.6 mL of water was distilled off azeotropically via a Dean–Stark trap. After cooling, about 20 mL of methanol were added and the precipitated diimide–diacid I [yield 68 g (90%)] was isolated by filtration and recrystallized from DMF as a yellow powder; mp 329°C.

ANAL. Calcd for $C_{45}H_{30}N_2O_{10}$ (758.7): C, 71.24%; H, 3.99%; N, 3.69%. Found: C, 70.67%; H, 4.22%; N, 3.94%.

Polymerization

A typical example of polymerization follows. A mixture of 1.8968 g (2.5 mmol) of diimide–diacid I, 0.5006 g (2.5 mmol) of 4,4′-oxydianiline (II_e), 1.6 g of calcium chloride, 1.61 g (5.2 mmol) of triphenyl phosphite, 4 mL of pyridine, and 16 mL of NMP was heated with stirring at 100°C for 3 h under nitrogen. The resulting polymer solution was trickled into 500 mL of methanol giving rise to a stringy, yellow precipitate which was washed thoroughly with methanol and hot water, collected by filtration, and dried. The yield was quantitative. Inherent viscosity of the polymer was 1.39 dL/g, measured on a concentration of 0.5 g/dL in DMAc–5% LiCl at 30°C.

Measurements

Poly(amide-imide) films were cast from the 10 g/dL of the DMAc solution of the obtained polymers. Inherent viscosities of the poly(amide-imide)s were determined at 30°C using 0.5 g/dL solutions in DMAc or DMAc–5% LiCl. A Canon–Fenske capillary viscometer was used without kinetic energy correction, since the solvent flow time exceeds 100 s. Weight-average molecular weight ($\bar{M}_w$) and number-average molecular weight ($\bar{M}_n$) were determined by gel permeation chromatography (GPC) on the basis of polystyrene calibration on a Shimadzu LC-6A apparatus (column: Tosoh TSK-GEL; eluent: DMF containing 0.01 mol/L of lithium bromide). IR spectra were recorded on a Jasco FT/IR-7000 Fourier transform infrared spectrometer. Elemental analyses were carried out with a Perkin-Elmer 240 C microanalyzer at Taiwan University (Taipei). Thermogravimetry (TG) was done on a Rigaku
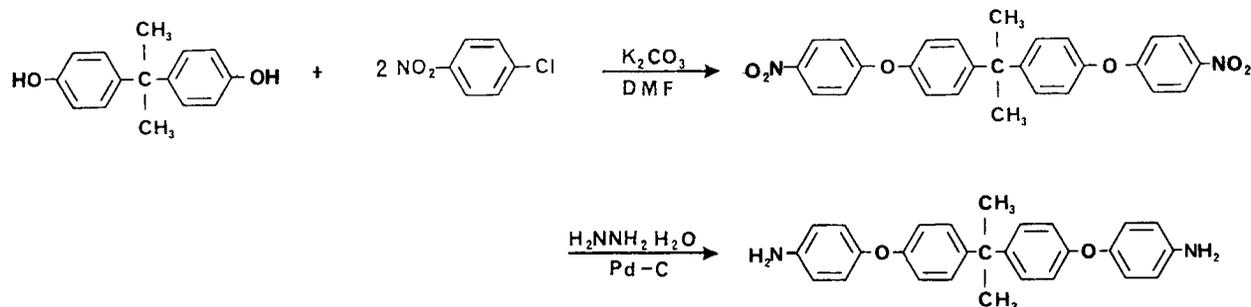
Scheme 1.

Thermoflex TG 8110 coupled to a Rigaku Thermal Analysis Station 100 on 10 ± 2 mg samples heated in flowing nitrogen (50 cm³/min) at a heating rate of 20°C/min. DSC was performed on a Sinku Riko TA-7000 thermal analysis controller at the rate of 20°C/min in flowing nitrogen (30 cm³/min). Wide angle x-ray diffractograms were obtained on a Rigaku Geiger D-Max III$_a$, using Ni-filtered CuK$_\alpha$ radiation (40 kV, 15 mA). An Instron Universal Tester Model 1130 with a load cell 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 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

Syntheses

2,2-Bis[4-(4-aminophenoxy)phenyl]propane (**II$_n$**) was prepared by reducing the dinitro compound obtained by condensation of bisphenol A with p-chloronitrobenzene (Scheme 1). 2,2-Bis[4-(4-trimellitimidophenoxy)phenyl]propane (**I**), the monomer with pre-formed imide rings, was synthesized via the two-stage procedure that included ring-opening addition of the diamine **II$_n$** with two equivalent amounts of trimellitic anhydride, followed by cyclodehydration to the imidodicarboxylic acid by means of toluene–water azeotropic distillation. The FT-IR spectrum of the dicarboxylic acid **I** is shown in Figure 1 (A).

A series of novel poly(amide-imide)s (**III$_{a-o}$**) containing isopropylidene and arylene ether units were prepared from diimide–diacid **I** and various aromatic diamines **II$_{a-o}$** by the direct polycondensation reaction using triphenyl phosphite and pyridine as condensing agents (Scheme 2).[9]

Almost all the reactions proceeded readily in ho-

mogeneous solution. The results of the polycondensation are summarized in Table I. These polymers were obtained in almost quantitative yields with inherent viscosities of 0.53–1.68 dL/g. The GPC curves indicated that $\bar{M}_w$ values of the poly(amide-imide)s were in the range of 89,000–206,900 relative to standard polystyrene, and the $\bar{M}_w/\bar{M}_n$ values, a measure of molecular weight distribution, were in the range of 1.90–2.46. The molecular weights of these polymers are sufficiently high to permit casting tough films.

The composition and structures of these polymers were confirmed by their elemental analyses and IR spectra. The elemental analysis values of these polymers are listed in Table II. In all cases, however,
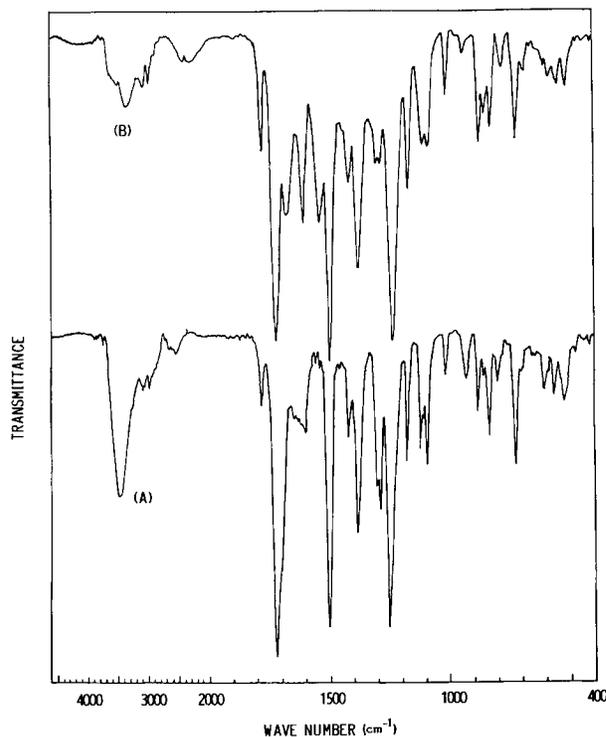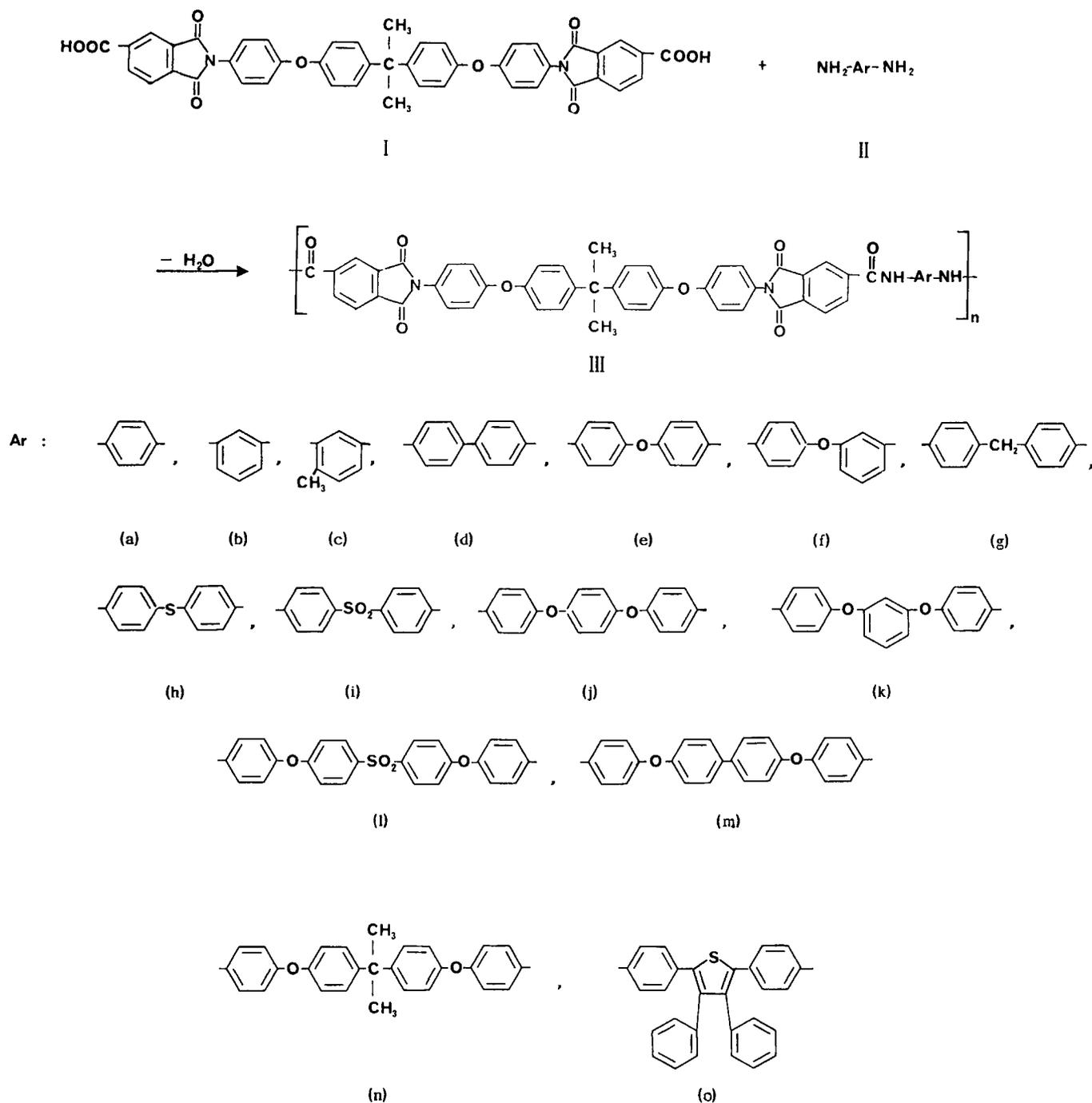

Figure 1. FT-IR spectra of: (A) diimide–diacid **I** (KBr) and (B) poly(amide-imide) **III$_b$** (film).

Scheme 2.

the found values of carbon were lower than the calculated values for the proposed structures. These may be attributed to the hygroscopic characteristics of amide group. The moisture intake for poly(amide-imide)s III_{a-o} was in the range of 2.02–7.14% at room temperature (ca. 23°C) 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 representative FT-IR spectrum of poly(amide-imide) III_b is shown in Figure 1(B). The IR spectra showed characteristic absorptions for the imide ring at 1779 and 1723 cm^{-1}, peculiar to the asymmetrical and symmetrical carbonyl stretching vibration, and at 1093 and 725 cm^{-1}, due to ring deformation. Bands of amide groups appear at 3352, 1678, and 1607 cm^{-1}.

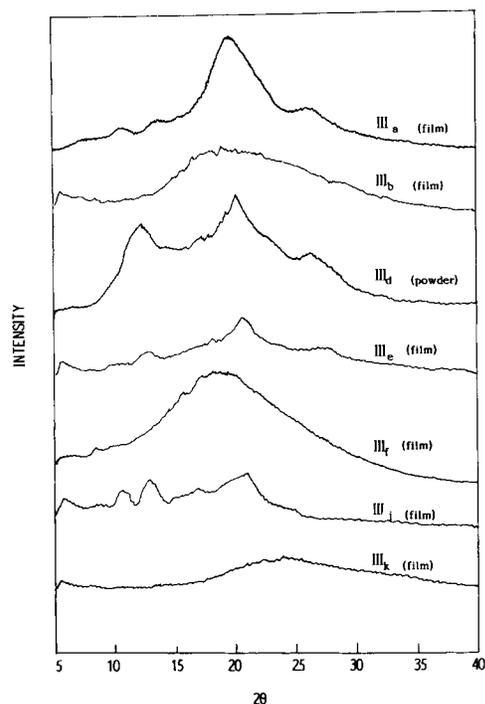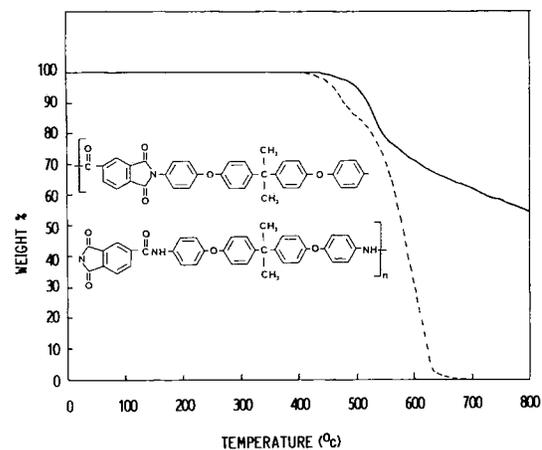**Table I.** Inherent Viscosities and Molecular Weights of Poly(amide-imide)s[a]

Polymer	η_{inh}[b] (dL/g)	$\bar{M}_w$[c] ($\times 10^{-4}$)	$\bar{M}_n$[c] ($\times 10^{-4}$)	$\bar{M}_w/\bar{M}_n$
III$_a$	1.12[d]	—	—	—
III$_b$	0.87	14.11	6.15	2.30
III$_c$	0.85	17.63	7.40	2.38
III$_d$	—	—	—	—
III$_e$	1.39[d]	—	—	—
III$_f$	0.86	19.51	8.08	2.41
III$_g$	0.77	11.55	5.28	2.19
III$_h$	0.82	17.58	7.13	2.46
III$_i$	0.53	8.93	4.65	1.90
III$_j$	1.52[d]	—	—	—
III$_k$	0.92	17.62	8.35	2.11
III$_l$	0.94	20.22	9.92	2.04
III$_m$	1.68[d]	—	—	—
III$_n$	1.24	20.69	8.91	2.32
III$_o$	1.03	17.37	7.81	2.23

[a] Polymerization was carried out using 2.5 mmol of diimide-diacid I and 2.5 mmol of diamine II with 5.2 mmol of triphenyl phosphite in 3 mL of pyridine and 16 mL of NMP containing 1.6 g of calcium chloride at 100°C for 3 h.
[b] Measured at 30°C on a concentration of 0.5 g/dL in DMAc.
[c] Determined by GPC.
[d] Measured at 30°C on a concentration of 0.5 g/dL in DMAc–5% LiCl.

Figure 3. TG curves of poly(amide-imide)s III_n with a heating rate of 20°C/min in (——) nitrogen and (- - - - -) air.

Properties of Polymers

The qualitative solubility of the poly(amide-imide)s in various solvents is shown in Table III. Most polymers were soluble in aprotic polar solvents such as DMAc, DMF, and NMP, and even in less polar solvents like m-cresol, o-chlorophenol, and pyridine; however, polymers III_d and III_m with biphenylene structure had somewhat limited solubility and were insoluble in all the tested solvents. This may be due to the semi-crystalline nature of these two polymers. All the polymers are insoluble in common organic solvents such as tetrahydrofuran, chloroform, acetone, and benzene. The superior solubility can be apparently attributed to the combined favorable effects of aromatic ether and isopropylidene linkages in the trimellitimide-based poly(amide-imide) backbone.

X-ray diffractograms of some representative polymers are shown in Figure 2. Most of the

Figure 2. Wide-angle x-ray diffractograms of some poly(amide-imide)s.
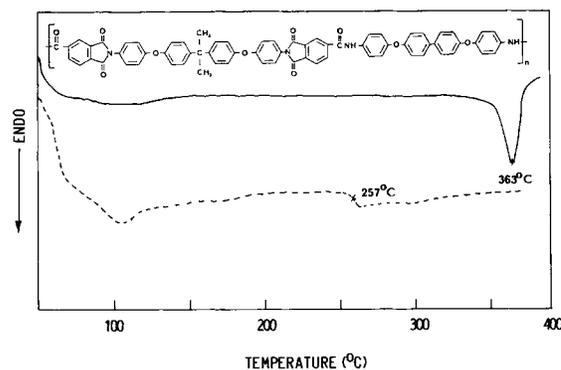
Figure 4. DSC thermograms of poly(amide-imide) III_m: (——) first heating, (- - -) second heating.

Table II. Elemental Analyses of Poly(amide-imide)s

Polymer	Formula (Molecular Weight)		Elemental Analysis (%)			Moisture Intake[a] (%)
			C	H	N	
III_a	$(C_{51}H_{34}N_4O_8)_n$	Calcd	73.73	4.12	6.74	
	$(830.85)_n$	Found	71.26	4.27	6.50	3.31
		Corrected[b]	73.62	4.13	6.72	
III_b	$(C_{51}H_{34}N_4O_8)_n$	Calcd	73.73	4.12	6.74	
	$(830.85)_n$	Found	71.22	4.30	6.48	3.47
		Corrected[b]	73.69	4.15	6.71	
III_c	$(C_{52}H_{36}N_4O_8)_n$	Calcd	73.92	4.29	6.63	
	$(844.88)_n$	Found	71.17	4.48	6.38	3.84
		Corrected[b]	73.90	4.31	6.62	
III_d	$(C_{57}H_{38}N_4O_8)_n$	Calcd	75.49	4.22	6.18	
	$(906.95)_n$	Found	73.21	4.37	5.97	3.07
		Corrected[b]	75.46	4.24	6.15	
III_e	$(C_{57}H_{38}N_4O_9)_n$	Calcd	74.18	4.15	6.07	
	$(922.95)_n$	Found	72.05	4.31	5.90	2.94
		Corrected[b]	74.17	4.18	6.07	
III_f	$(C_{57}H_{38}N_4O_9)_n$	Calcd	74.18	4.15	6.07	
	$(922.95)_n$	Found	71.91	4.29	5.87	3.13
		Corrected[b]	74.16	4.16	6.05	
III_g	$(C_{58}H_{40}N_4O_8)_n$	Calcd	75.64	4.38	6.08	
	$(920.98)_n$	Found	73.32	4.54	5.87	3.14
		Corrected[b]	75.62	4.40	6.05	
III_h	$(C_{57}H_{38}N_4O_8S)_n$	Calcd	72.91	4.08	5.97	
	$(939.02)_n$	Found	70.53	4.26	5.77	3.33
		Corrected[b]	72.88	4.12	5.96	
III_i	$(C_{57}H_{38}N_4O_{10}S)_n$	Calcd	70.51	3.94	5.77	
	$(971.02)_n$	Found	68.21	4.14	5.55	3.36
		Corrected[b]	70.50	4.00	5.74	
III_j	$(C_{63}H_{42}N_4O_{10})_n$	Calcd	74.55	4.17	5.52	
	$(1015.05)_n$	Found	72.81	4.40	5.37	2.35
		Corrected[b]	74.52	4.20	5.50	
III_k	$(C_{63}H_{42}N_4O_{10})_n$	Calcd	74.55	4.17	5.52	
	$(1015.05)_n$	Found	72.81	4.29	5.36	2.36
		Corrected[b]	74.53	4.19	5.49	
III_l	$(C_{69}H_{46}N_4O_{12}S)_n$	Calcd	71.74	4.01	4.85	
	$(1155.21)_n$	Found	69.76	4.16	4.68	2.80
		Corrected[b]	71.71	4.04	4.81	
III_m	$(C_{69}N_{46}N_4O_{10})_n$	Calcd	75.95	4.25	5.13	
	$(1091.15)_n$	Found	74.28	4.37	5.00	2.34
		Corrected[b]	75.94	4.27	5.12	
III_n	$(C_{72}H_{52}N_4O_{10})_n$	Calcd	76.31	4.63	4.94	
	$(1133.23)_n$	Found	74.78	4.74	4.82	2.02
		Corrected[b]	76.29	4.64	4.92	
III_o	$(C_{73}H_{48}N_4O_8S)_n$	Calcd	76.83	4.24	4.91	
	$(1141.27)_n$	Found	71.66	4.62	4.55	7.14
		Corrected[b]	76.78	4.29	4.87	

[a] Moisture intake (%) $= \dfrac{W - W_o}{W_o} \times 100\%$.

W = weight of polymer sample after standing at room temperature, and W_o = weight of polymer sample after dried in vacuum at 100°C for 10 h.

[b] For C and N: Corrected value = found value $\times$ (1 + moisture intake %).

For H: Corrected value = found value $\times$ (1 − moisture intake %).

Table III. Solubility of Poly(amide-imide)s[a]

Polymer	Solvent[b]						
	DMAc	DMF	NMP	Py	m-Cresol	o-Chlorophenol	DMAc–5% LiCl
III_a	−−	−−	++	−−	−−	−−	++
III_b	++	++	++	++	++	++	++
III_c	++	++	++	++	++	++	++
III_d	−−	−−	−−	−−	−−	−−	−−
III_e	+−	−−	++	−−	−−	−−	++
III_f	++	+−	++	++	++	++	++
III_g	++	+−	++	−−	++	++	++
III_h	++	++	++	++	+−	++	++
III_i	++	++	++	++	++	++	++
III_j	−−	−−	++	−−	−−	−−	++
III_k	++	++	++	++	++	++	++
III_l	++	++	++	++	++	++	++
III_m	−−	−−	−−	−−	−−	−−	++
III_n	++	++	++	++	++	++	++
III_o	++	++	++	++	++	++	++

[a] Solubility: ++, soluble; +−, partially or swelling; −−: insoluble at room temperature.
[b] DMF, dimethylformamide; DMAc, dimethylacetamide; NMP, N-methyl-2-pyrrolidone; Py, pyridine.

poly(amide-imide)s derived from imide-containing dicarboxylic acid **I**, except polymers **III_a, d, e, and j**, exhibit amorphous patterns. The semi-crystalline behavior of the polymers **III_a, d, e, and j** may be attributed to their rigid structure and caused the insoluble nature in polar solvents as mentioned above, which is in agreement with the general rule that the solubility decreases with increasing crystallinity.

Transparent, flexible, and tough films could be cast from the DMAc solutions of most of the obtained poly(amide-imide)s. The tensile properties of some typical poly(amide-imide)s are summarized in Table IV. The films had tensile strength of 73–107 MPa, elongation to break of 6–90%, and initial modulus of 1.67–2.34 GPa. Except for the brittle behavior of polymer **III_a**, all the polymers listed in Table IV behave as ductile materials with good tensile strengths and moderate elongations-to-break.

The thermal stability of the poly(amide-imide) was studied by thermogravimetric analysis (TG). Figure 3 shows typical TG curves of poly(amide-imide) **III_n**. The temperatures of 10% weight loss in nitrogen and air atmosphere were determined from original thermograms and tabulated in Table V. In general, all the polymers exhibited good thermal stability with no significant weight loss up to temperatures of ca. 450°C in nitrogen, and their 10% weight loss temperatures were recorded in the range of 508–538°C in nitrogen and 473–499°C in air, respectively (Table V).

Table V also summarizes the thermal transition data of the poly(amide-imide)s. Figure 4 shows

typical DSC curves of polymer **III_m**. Quenching from the elevated temperatures to room temperature in air 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 were in the range of 237–293°C. Although some as-precipitated polymers (such as polymers **III_a, d, e, j, and m**) melted, they failed to crystallize under the conditions of the DSC measurement and exhibited clear T_gs in second heating runs as shown in Figure 4. No cold crystallization was observed above T_g. Thus, it ap-

Table IV. Tensile Properties of Poly(amide-imide) Films

Polymer[a]	Yield Strength (MPa)	Tensile Strength (MPa)	Elongation at Break (%)	Tensile Modulus (GPa)
III_b	78	73	39	2.10
III_c	85	75	31	2.13
III_e[b]	—	101	6	2.05
III_f	73	79	90	1.98
III_i	—	82	6	2.18
III_j[b]	—	107	6	2.01
III_k	70	74	83	1.67
III_l	78	95	77	1.76
III_n	85	88	53	2.34
III_o	—	82	38	2.14

[a] Films were cast from polymer solutions in DMAc.
[b] Films were cast from polymer solutions in NMP.

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

| Polymer | DSC | | TG | |
| | $T_g{}^a$ (°C) | $T_m{}^b$ (°C) | Decomposition Temperature^c (°C) | |
			In Air	In Nitrogen
III$_a$	257	340	523	520
III$_b$	278	—	478	511
III$_c$	268	—	480	512
III$_d$	263	388	499	520
III$_e$	258	325	480	531
III$_f$	251	—	485	518
III$_g$	245	—	473	517
III$_h$	267	—	489	508
III$_i$	263	—	497	515
III$_j$	248	325	487	525
III$_k$	237	—	492	515
III$_l$	265	—	487	522
III$_m$	257	363	475	523
III$_n$	244	—	478	519
III$_o$	293	—	489	538

^{a} From the second heating traces of DSC measurements conducted with a heating rate of 20°C/min in nitrogen.

^{b} Temperatures of the endotherm peaks in the first heating trace of DSC. All the peaks disappeared in the second heating DSC trace after quenching from 400°C to room temperature in air.

^{c} Temperatures at which 10% weight loss was recorded by TG at heating rate of 20°C/min.

pears that the initial crystallinity results from the presence of solvent and the way these material were precipitated. Sharp melting (T_m) endotherm peaks of polymers III$_{a, d, e, j, and m}$ could be observed on their first DSC traces.

CONCLUSIONS

The diimide–diacid 2,2-bis[4-(4-trimellitimido-phenoxy)phenyl]propane was used as a poly(amide-imide) building block. Poly(amide-imide)s having moderate to high molecular weight were successfully prepared by direct polycondensation of the diimide–diacid with various aromatic diamines by means of triphenyl phosphite and pyridine. Polymers, in general, showed good thermal stability and excellent solubility, and the casting films exhibited excellent tensile properties. Thus, the present poly(amide-imide)s are considered to be new candidates for processable high-performance polymeric materials.

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