

# New Poly(amide-Imide)s Syntheses. II. Soluble Poly(amide-Imide)s Derived from 2,5-Bis(4-Aminophenyl)-3,4-Diphenylthiophene and Various *N*-( $\omega$ -Carboxyalkyl)-trimellitimidides, *N*-(Carboxyphenyl)trimellitimidides, or *N,N'*-Bis( $\omega$ -Carboxyalkyl)pyromellitimidides

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## SYNOPSIS

Various dicarboxylic acids with preformed imide rings were readily obtained by the condensation of  $\omega$ -amino acids and aminobenzoic acids with trimellitic anhydride, and  $\omega$ -amino acids with pyromellitic dianhydride. New tetraphenylthiophene-containing poly(amide-imide)s having inherent viscosities of 0.58–1.54 dL/g were prepared by the direct polycondensation reaction of 2,5-bis(4-aminophenyl)-3,4-diphenylthiophene with the imide-containing dicarboxylic acids using triphenyl phosphite and pyridine as condensing agents. These polymers were amorphous and readily soluble in a variety of solvents such as dimethylacetamide (DMAc), dimethylformamide (DMF), and *N*-methyl-2-pyrrolidone (NMP), and could be easily solution cast into transparent, flexible, and tough films. Most of the poly(amide-imide)s showed clear glass transition on the heating traces of differential scanning calorimetry (DSC) in the range of 146–244°C. Almost all the poly(amide-imide)s exhibited no appreciable decomposition below 400°C, with 10% weight loss being recorded above 420°C in nitrogen. © 1992 John Wiley & Sons, Inc.

**Keywords:** poly(amide-imide)s • pyromellitimidides • tetraphenylthiophene diamine • trimellitimidides • triphenyl phosphite

## INTRODUCTION

Recent intensive investigations in the field of high-temperature polymers have resulted in the development of numerous classes of thermally stable polymers. The unusual thermal resistance of these polymers are due to the presence of aromatic and heterocyclic rings along the backbone connected by thermally stable bonds. However, a wide variety of these materials are difficult to fabricate because of their low solubility in organic solvents and high melting or softening temperatures. For this purpose, several approaches such as introduction of bulky pendant groups in the main chain, placing flexible

bondings on the backbone or synthesis of cardo polymer<sup>1</sup> have been taken to obtain processable or soluble polymer.

In recent years, Imai and co-workers<sup>2–10</sup> have developed soluble high-temperature polymers, such as polyimides, poly(amide-imide)s, polyesters, poly(ester-imide)s, and polyazomethines, which possess a highly phenylated tetraphenylthiophene unit as the key structure. The poly(amide-imide)s reported were prepared by the reaction of tetraphenylthiophene diamine with 4-chloroformylphthalic anhydride, and tetraphenylthiophene diisocyanate with trimellitic anhydride. However, the inherent viscosities reported were not very high. Recently, the reaction promoted by triphenyl phosphite<sup>11</sup> as a condensing agent was successfully applied to the direct preparation of poly(amide-imide)s<sup>12–15</sup> from imide-containing dicarboxylic acids

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and aromatic diamines in the mixture of pyridine and amide solvents such as *N*-methyl-2-pyrrolidone (NMP) in the presence of metal salts such as calcium chloride, giving polymers of high molecular weights. The purpose of this research was to synthesize poly(amide-imide)s from 2,5-bis(4-aminophenyl)-3,4-diphenylthiophene with various trimellitimide- or pyromellitimide-containing dicarboxylic acids by means of triphenyl phosphite. The solubility, tensile properties, thermal properties, and crystallinity of the resultant novel poly(amide-imide)s are also described.

## EXPERIMENTAL

### Materials

Pyromellitic dianhydride (Fluka) was recrystallized from boiling acetic anhydride. Trimellitic anhydride (Wako) was used without previous purification. Amino acids (TCI) were used without any further purification. Commercially obtained anhydrous LiCl and CaCl<sub>2</sub> were dried under vacuum at 150°C for 6 h and 180°C for 10 h, respectively. NMP and pyridine were purified by distillation under reduced pressure over calcium hydride and stored over 4 Å molecular sieves. Triphenyl phosphite (TCI) was purified by distillation under reduced pressure. The imide-dicarboxylic acids **II**<sub>a-f</sub> and **IV**<sub>a-g</sub> were obtained as previously reported,<sup>12,15</sup> from the condensation of amino acids with trimellitic anhydride and pyromellitic dianhydride. According to the reported method,<sup>2</sup> 2,5-bis(4-aminophenyl)-3,4-diphenylthiophene (**I**) was synthesized starting from tetraphenylthiophene, which was easily obtained by the reaction of benzyl chloride with powdered sulfur at an elevated temperature.

### Polymerization

A typical example of polymerization was as follows.

#### **Polymer V<sub>f</sub> from Diamine I and Diimide-Diacid IV<sub>f</sub>**

A mixture of 0.730 g (1.25 mmol) of diacid **IV**<sub>f</sub>, 0.522 g (1.25 mmol) of 2,5-bis(4-aminophenyl)-3,4-diphenylthiophene (**I**), 0.3 g of calcium chloride, 0.6 g of lithium chloride, 0.8 mL (2.5 mmol) of triphenyl phosphite, 1.6 mL of pyridine, and 7 mL of NMP was heated with stirring at 100°C for 3 h under nitrogen. The obtained polymer solution was trickled on 500 mL of methanol giving rise to a stringy precipitate which was washed thoroughly with meth-

anol and hot water, collected by filtration, and dried. The yield was quantitative. The inherent viscosity of a 0.5 g/dL polymer solution in DMAc was 1.54 dL/g at 30°C.

IR (film): 1773 and 1723 (C=O imide), 1075 and 729 (imide ring), 1671 (C=O amide), 3314 (N—H amide) cm<sup>-1</sup>.

ANAL. Calcd for (C<sub>60</sub>H<sub>62</sub>N<sub>4</sub>O<sub>6</sub>S · H<sub>2</sub>O)<sub>n</sub>: C, 73.14; H, 6.45; N, 5.69. Found: C, 73.55; H, 6.41; N, 5.77.

All other poly(amide-imide)s were synthesized by an analogous procedure as above.

### Preparation of Poly(amide-Imide) Films

About 10 wt % solutions of poly(amide-imide)s in DMAc or NMP were cast onto glass substrate at 80°C. The semi-dried polymer films were removed from the substrate and then heated at 120°C for 8 h in vacuum. All films were transparent and approximately 0.1 mm in thickness. These poly(amide-imide) films were cut into strips of 6 cm length and 0.5 cm width for stress-strain measurements.

### Measurements

Elemental analyses were run in a Perkin-Elmer Model 240 C, H, N analyzer. IR spectra were recorded on a Jasco FT/IR-7000 Fourier-transform Infrared Spectrometer. The inherent viscosities were measured with a Cannon-Fenske viscosimeter thermostated at 30°C. Weight-average molecular weight ( $\bar{M}_w$ ) and number-average molecular weight ( $\bar{M}_n$ ) were determined by means of 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).

The DSC traces were measured on a Dupont 910 differential scanning calorimeter coupled to a Dupont 1090 thermal analyzer at the rate of 20°C/min in flowing nitrogen (30 cm<sup>3</sup>/min). Thermogravimetry (TG) was conducted with a Dupont 951 thermogravimetric analyzer coupled to a Dupont 1090 thermal analyzer. Experiments were carried out on 10 ± 2 mg samples heated in flowing nitrogen (50 cm<sup>3</sup>/min) at a heating rate of 20°C/min.

The wide-angle x-ray measurements were performed at room temperature (about 25°C) with film specimens of about 0.1 mm thick on a Rigaku Geiger Flex D-Max III<sub>a</sub> x-ray diffractometer, using Ni-filtered CuK<sub>α</sub> radiation (40 kV, 15 mA). The scanning rate was 2°/min.

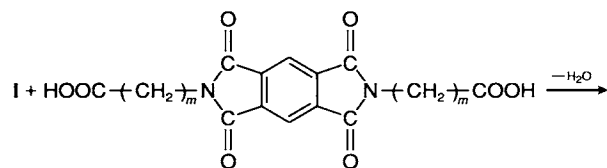
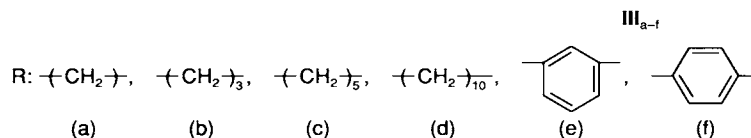
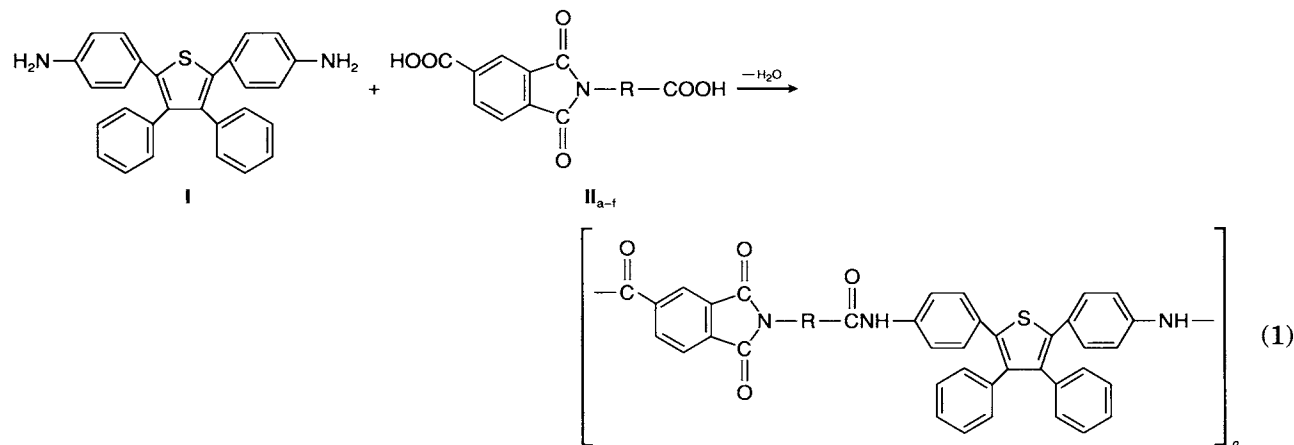
An Instron Universal Tester Model 1130 with a load cell 5 kg was used to study the stress-strain behavior of the samples. A gauge of 2 cm and a strain rate of 5 cm/min were used for this study. Measurements were performed at room temperature with film specimens (0.5 cm wide, 6 cm long, and about 0.1 mm thick) and an average of at least five individual determinations was used.

## RESULTS AND DISCUSSION

### Polymer Synthesis

Two series of dicarboxylic acids having preformed imide rings, **II**<sub>a-f</sub> and **IV**<sub>a-g</sub>, were synthesized for use in this study by the condensation of trimellitic anhydride and pyromellitic dianhydride with the cor-

responding amino acids. Characterization data of these imide-containing dicarboxylic acids were described previously.<sup>12,13,15</sup> The tetraphenylthiophene diamine **I** of the structure as shown in eq. (1) was prepared starting from benzyl chloride and sulfur by the synthetic route reported by Imai et al.<sup>2</sup> The poly(amide-imide)s containing terphenylthiophene units were prepared [eqs. (1) and (2)] by the direct polycondensation technique using tetraphenylthiophene-containing diamine **I** with structurally different imidodicarboxylic acids by the use of triphenyl phosphite (TPP) and pyridine as condensing agent. Table I summarizes the results of the polymerizations. Almost all the poly(amide-imide)s were obtained quantitatively, with moderate to high inherent viscosities in the range of 0.58–1.54 dL/g. The GPC curves indicated that  $\bar{M}_n$  values of the



**IV**<sub>a-g</sub>:  $m = 1, 2, 3, 4, 5, 10, 11$

**V**<sub>a-g</sub>:  $m = 1, 2, 3, 4, 5, 10, 11$

**Table I.** Synthesis of Poly(amide-Imide)s<sup>a</sup>

Polymer	$\eta_{inh}^b$ (dL/g)	$\bar{M}_w^d$ ( $\times 10^{-4}$ )	$\bar{M}_n^d$ ( $\times 10^{-4}$ )	$\bar{M}_w/\bar{M}_n$
III <sub>a</sub>	0.81	16.2	10.2	1.6
III <sub>b</sub>	0.71	10.4	6.6	1.6
III <sub>c</sub>	0.83	13.2	9.5	1.4
III <sub>d</sub>	0.93	21.6	12.6	1.7
III <sub>e</sub>	0.58	14.7	9.6	1.5
III <sub>f</sub>	1.43 <sup>c</sup>	—	—	—
V <sub>a</sub>	0.77	23.5	11.8	2.0
V <sub>b</sub>	1.49	34.4	15.5	2.2
V <sub>c</sub>	0.72	22.2	13.5	1.6
V <sub>d</sub>	1.30	38.7	16.8	2.3
V <sub>e</sub>	0.68	12.4	8.4	1.5
V <sub>f</sub>	1.54	39.7	19.3	2.0
V <sub>g</sub>	1.07	27.3	14.1	1.9

<sup>a</sup> Polymerization was carried out with 1.25 mmol of each monomer, 0.8 mL of TPP, 1.6 mL of pyridine, 0.3 g of CaCl<sub>2</sub>, and 0.6 g of LiCl in 7 mL of NMP at 100°C for 3 h under nitrogen.

<sup>b</sup> Measured at 30°C on a concentration of 0.5 g/dL in DMAc.

<sup>c</sup> Measured in DMAc containing 5 wt % of LiCl dissolved.

<sup>d</sup> Determined by GPC.

poly(amide-imide)s were in the range of 66,000–168,000 relative to standard polystyrene, and the  $\bar{M}_w/\bar{M}_n$ , a measure of molecular weight distribution, were rather low (1.4–2.3).

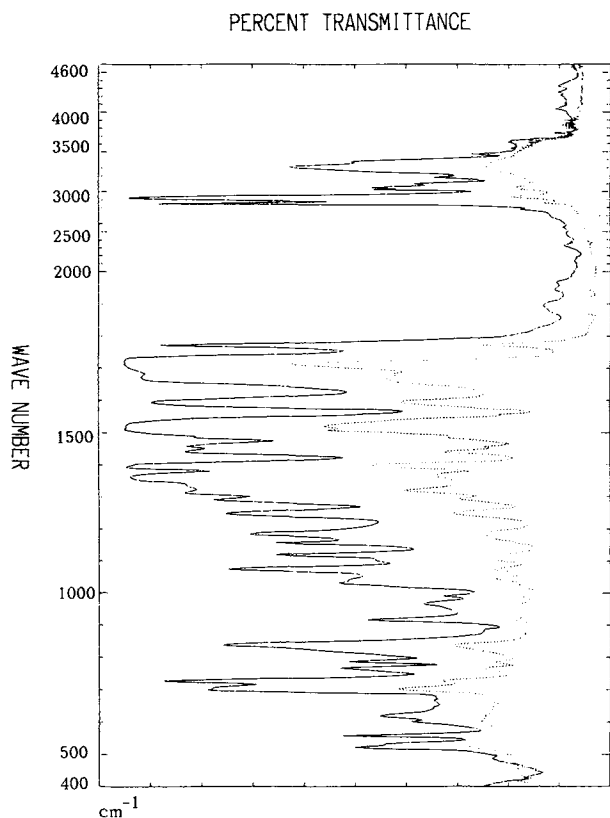
The elemental analysis values of these polymers were in good agreement with the calculated values for the proposed structures, assuming that the poly(amide-imide)s absorbed one to two molecules of H<sub>2</sub>O per repeating unit (Table II). Figure 1 shows the representative FTIR spectra of polymers III<sub>d</sub> and V<sub>f</sub>. The IR spectra exhibited characteristic absorptions for the imide ring at 1775 and 1715 cm<sup>-1</sup>, peculiar to the asymmetrical and symmetrical carbonyl stretching vibration, and at 1075 and 730 cm<sup>-1</sup> due to ring deformation. Bands of amide groups appear at 3315, 1670, and 1595 cm<sup>-1</sup>.

### Properties of Polymers

The qualitative solubility of poly(amide-imide)s in various solvents is shown in Table III. Almost all poly(amide-imide)s having the tetraphenylthio-

**Table II.** Elemental Analysis of Poly(amide-Imide)s

Polymer	Formula (Molecular Weight)	C (%)		H (%)		N (%)	
		Calcd	Found	Calcd	Found	Calcd	Found
III <sub>a</sub>	(C <sub>39</sub> H <sub>25</sub> N <sub>3</sub> O <sub>4</sub> S · 1.5H <sub>2</sub> O) <sub>n</sub> (658.74) <sub>n</sub>	71.11	71.38	4.07	3.98	6.38	6.35
III <sub>b</sub>	(C <sub>41</sub> H <sub>29</sub> N <sub>3</sub> O <sub>4</sub> S · H <sub>2</sub> O) <sub>n</sub> (677.78) <sub>n</sub>	72.66	72.83	4.61	4.31	6.20	6.19
III <sub>c</sub>	(C <sub>43</sub> H <sub>33</sub> N <sub>3</sub> O <sub>4</sub> S · 1.5H <sub>2</sub> O) <sub>n</sub> (714.85) <sub>n</sub>	72.25	72.35	4.86	4.68	5.88	5.86
III <sub>d</sub>	(C <sub>48</sub> H <sub>43</sub> N <sub>3</sub> O <sub>4</sub> S · H <sub>2</sub> O) <sub>n</sub> (775.97) <sub>n</sub>	74.30	74.75	5.85	5.63	5.32	5.50
III <sub>e</sub>	(C <sub>44</sub> H <sub>27</sub> N <sub>3</sub> O <sub>4</sub> S · 1.5H <sub>2</sub> O) <sub>n</sub> (720.81) <sub>n</sub>	73.32	73.34	3.98	3.81	5.83	5.82
III <sub>f</sub>	(C <sub>44</sub> H <sub>27</sub> N <sub>3</sub> O <sub>4</sub> S · 1.5H <sub>2</sub> O) <sub>n</sub> (720.81) <sub>n</sub>	73.32	73.57	3.98	3.96	5.83	5.93
V <sub>a</sub>	(C <sub>42</sub> H <sub>26</sub> N <sub>4</sub> O <sub>6</sub> S · 1.5H <sub>2</sub> O) <sub>n</sub> (741.79) <sub>n</sub>	68.01	68.84	3.74	3.85	7.55	7.65
V <sub>b</sub>	(C <sub>44</sub> H <sub>30</sub> N <sub>4</sub> O <sub>6</sub> S · 2H <sub>2</sub> O) <sub>n</sub> (778.81) <sub>n</sub>	67.86	67.93	3.88	3.91	7.19	7.05
V <sub>c</sub>	(C <sub>46</sub> H <sub>34</sub> N <sub>4</sub> O <sub>6</sub> S · 2H <sub>2</sub> O) <sub>n</sub> (806.89) <sub>n</sub>	68.47	68.63	4.75	4.35	6.94	7.00
V <sub>d</sub>	(C <sub>48</sub> H <sub>38</sub> N <sub>4</sub> O <sub>6</sub> S · 1.5H <sub>2</sub> O) <sub>n</sub> (825.94) <sub>n</sub>	69.80	69.79	4.82	4.57	6.78	6.58
V <sub>e</sub>	(C <sub>50</sub> H <sub>42</sub> N <sub>4</sub> O <sub>6</sub> S · 1.5H <sub>2</sub> O) <sub>n</sub> (854.00) <sub>n</sub>	70.32	70.64	5.31	5.16	6.56	6.60
V <sub>f</sub>	(C <sub>60</sub> H <sub>62</sub> N <sub>4</sub> O <sub>6</sub> S · H <sub>2</sub> O) <sub>n</sub> (985.26) <sub>n</sub>	73.14	73.55	6.45	6.41	5.69	5.77
V <sub>g</sub>	(C <sub>62</sub> H <sub>66</sub> N <sub>4</sub> O <sub>6</sub> S · H <sub>2</sub> O) <sub>n</sub> (1013.31) <sub>n</sub>	73.49	73.79	6.66	6.54	5.53	5.58



**Figure 1.** FTIR spectra of poly(amide-imide)s **III<sub>d</sub>** (-----) and **V<sub>f</sub>** (—).

phene unit were highly soluble in a variety of organic solvents including amide-type solvents, DMSO, *m*-cresol, and pyridine; but were insoluble in tetrahy-

drofuran, chloroform, acetone, and benzene. The excellent solubility behavior may be attributed to the incorporation of the bulky pendant groups of tetraphenylthiophene unit into the polymer backbones. However, the introduction of *p*-phenylene structure into the polymer backbone decreases solubility of the polymers as in the case of **III<sub>f</sub>**. These poly(amide-imide)s based on tetraphenylthiophene diamine show apparently better solubility than their homologs with other diamine moieties as described in our previous report.<sup>12,15</sup>

Strong and flexible films could be cast from the DMAc or NMP solution of most of the obtained poly(amide-imide)s. The x-ray diffraction studies indicated that all the poly(amide-imide)s were amorphous, possibly due to the presence of bulky, pendant phenyl groups of the tetraphenylthiophene units. The tensile properties of some typical poly(amide-imide)s are summarized in Table IV. In tension, some polymer films such as those from **III<sub>a-e</sub>** and **V<sub>e,g</sub>** behave as a ductile material with a yield point and moderate elongation to break. The ductile nature of the polyamide-imide films may be due in part to the introduction of flexible methylene groups and *m*-phenylene group (such as in the case of polymer **III<sub>e</sub>**) into the polymer main chain. Polymer **III<sub>f</sub>**, possessing the rigid *p*-phenylene group and highly aromatic tetraphenylthiophene unit showed higher breakdown strength and initial modulus but lower elongation.

The thermal behavior of the polymers was evaluated by means of differential scanning calorimetry

**Table III.** Solubility of Poly(amide-Imide)s<sup>a</sup>

Polymer	Solvent <sup>b</sup>					
	DMAc	DMF	NMP	DMSO	Py	<i>m</i> -Cresol
<b>III<sub>a</sub></b>	+	+	+	+	+	+
<b>III<sub>b</sub></b>	+	+	+	+	+	+
<b>III<sub>c</sub></b>	+	+	+	+	+	+
<b>III<sub>d</sub></b>	+	+	+	+	+	+
<b>III<sub>e</sub></b>	+	+	+	+	+	+
<b>III<sub>f</sub></b>	-	-	+	-	-	-
<b>V<sub>a</sub></b>	+	+	+	+	+	+
<b>V<sub>b</sub></b>	+	+	+	-	-	-
<b>V<sub>c</sub></b>	+	+	+	+	+	+
<b>V<sub>d</sub></b>	+	+	+	+	+	+
<b>V<sub>e</sub></b>	+	+	+	+	+	+
<b>V<sub>f</sub></b>	+	+	+	-	+	+
<b>V<sub>g</sub></b>	+	+	+	-	+	+

<sup>a</sup> Solubility: +, soluble; -, insoluble.

<sup>b</sup> DMF, dimethylformamide; DMAc, dimethylacetamide; NMP, *N*-methyl-2-pyrrolidone; DMSO, dimethylsulfoxide; Py, pyridine.

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

Polymer <sup>a</sup>	Strength at Yield (MPa)	Strength at Break (MPa)	Elongation to Break (%)	Initial Modulus (GPa)
III <sub>a</sub>	63	57	52	1.53
III <sub>b</sub>	81	73	16	2.10
III <sub>c</sub>	46	37	56	1.61
III <sub>d</sub>	45	39	23	1.11
III <sub>e</sub>	60	53	17	1.60
III <sub>f</sub>	—	110	9	2.63
V <sub>b</sub> <sup>b</sup>	—	84	12	1.89
V <sub>d</sub>	—	67	30	1.73
V <sub>e</sub>	69	59	36	1.63
V <sub>f</sub>	—	52	56	1.24
V <sub>g</sub>	32	26	147	0.83

<sup>a</sup> Films were cast from polymer solutions of DMAc.<sup>b</sup> Cast from polymer solutions of NMP.

(DSC) and thermogravimetry (TG). With the exception of polymers III<sub>f</sub> and V<sub>a</sub>, the polymers displayed discernable base line shifts, i.e.,  $T_g$ 's, on their second heating traces of DSC measurements. For the wholly aromatic poly(amide-imide)s, the DSC thermogram of polymer III<sub>f</sub> derived from *N*-(*p*-carboxyphenyl)trimellitimide showed no discerna-

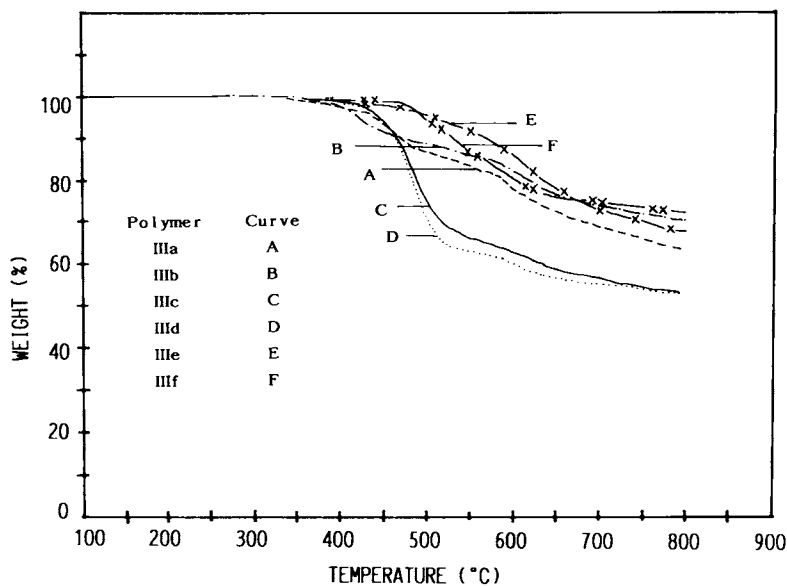
ble transition, whereas that of the *m*-isomer (III<sub>e</sub>) showed a clear  $T_g$  around 235°C. For the aliphatic-aromatic poly(amide-imide)s,  $T_g$ 's were high for those with short aliphatic chains and were lowered with increasing length of the methylene segment; the range of values was 186–216°C for III<sub>a-d</sub> and 146–244°C for V<sub>b-g</sub> (Table V). Polymer V<sub>a</sub> containing only one methylene in the repeating unit showed no discernable transition on the DSC curve. Well-defined melting points of the polymers could not be detected by DSC, due to their amorphous nature as evidenced by the x-ray diffraction measurements.

Thermogravimetric traces indicate that all the poly(amide-imide)s possess moderately high thermal stability with no significant weight loss up to temperatures of approximately 400°C in a nitrogen atmosphere (Figs. 2 and 3). As expected, wholly aromatic poly(amide-imide)s III<sub>e,f</sub> are found to be more stable than the aliphatic-aromatic ones. From the comparison among the TG curves of the alkylene-linked poly(amide-imide)s it is found that the introduction of longer linear aliphatic chains does not cause a decrease of initial decomposition temperature. The poly(amide-imide)s with 10 or 11 methylene units started losing weight at higher temperatures than those with 1 to 5 methylenes. The reason has been demonstrated in the previous report.<sup>15</sup> TG data, temperatures at various percents

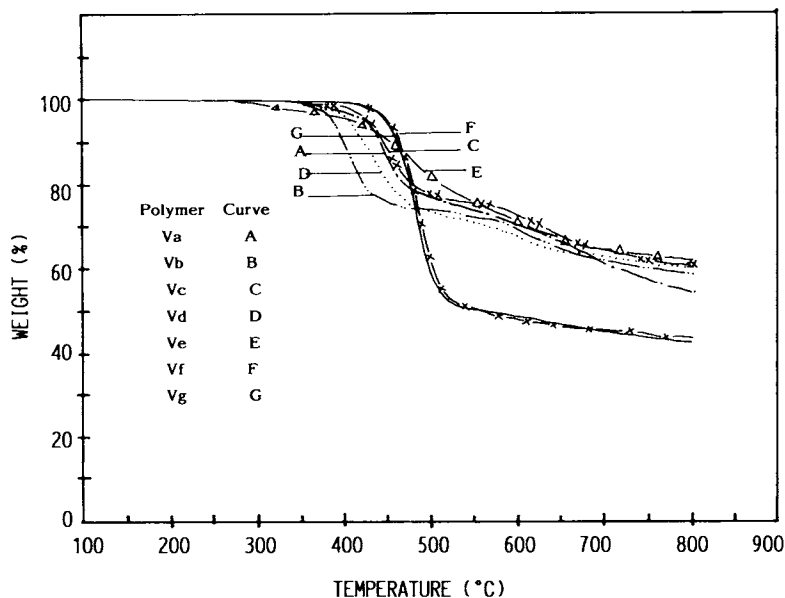
**Table V.** Thermal Behavior Data of Poly(amide-Imide)s

Polymer	DSC $T_g^a$ (°C)	TG <sup>b</sup>			
		Temperature (°C) at Various Wt. Loss			Wt % Residual at 800°C
		5%	10%	30%	
III <sub>a</sub>	216	436	469	673	65
III <sub>b</sub>	194	422	462	788	69
III <sub>c</sub>	190	447	465	519	53
III <sub>d</sub>	181	443	464	502	52
III <sub>e</sub>	235	500	563	743	62
III <sub>f</sub>	— <sup>c</sup>	502	532	860	71
V <sub>a</sub>	— <sup>c</sup>	423	442	599	54
V <sub>b</sub>	244	383	397	560	58
V <sub>c</sub>	242	424	444	617	60
V <sub>d</sub>	238	408	426	562	59
V <sub>e</sub>	206	409	453	611	61
V <sub>f</sub>	150	447	462	489	43
V <sub>g</sub>	146	445	460	487	42

<sup>a</sup> From the second heating traces of DSC measurements conducted with a heating rate of 20°C/min in nitrogen.<sup>b</sup> Thermogravimetry was conducted at a heating rate of 20°C/min in nitrogen.<sup>c</sup> Not observed before decomposition.



**Figure 2.** TG curves of poly(amide-imide)s III<sub>a-f</sub>, with a heating rate of 20°C/min in nitrogen.



**Figure 3.** TG curves of poly(amide-imide)s V<sub>a-g</sub>, with a heating rate of 20°C/min in nitrogen.

of weight loss and char yield at 800°C, of all polymers are also summarized in Table V.

In conclusion, these tetraphenylthiophene-containing poly(amide-imide)s could be easily processed into end-use products like films with excellent mechanical properties and moderately high thermal stability. Thus, they may be considered to be promising high-performance plastic materials.

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