

# Heat-Resistant Thermosetting Polyimide Matrix Resins Containing Keto, Ether, Hexafluoroisopropylidene Linkages and Maleimido- and Nadimido- End Capped Groups

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## SYNOPSIS

Various heat-resistant thermosetting polyimides containing hexafluoroisopropylidene and keto and ether groups suitable for fiber-reinforced composites development have been synthesized by thermal polymerization of maleimido and nadimido end capped polymer precursors. Thermal polymerization involving addition reactions was performed at 225°C for 1.5 h and post-curing at 290°C for 0.5 h. Tough polymers XVIII to XXIII were obtained. The thermal polymerization was monitored using infrared spectroscopy. The polymer precursors were characterized by IR, <sup>1</sup>H-NMR, and elemental analysis. The synthesized polymers were evaluated for thermal stability using dynamic thermogravimetric analysis (TGA). All the polymers showed thermal decomposition temperatures in the range of 430–435°C in nitrogen and in air. The char yield of the polymers was in the range of 60–67% in nitrogen at 800°C. This study indicated that thermally synthesized polymers from hexafluoroisopropylidene, keto, and ether containing polyimides are potential candidates for the development of advanced materials for aerospace and high-tech applications. © 1993 John Wiley & Sons, Inc.

## INTRODUCTION

Aromatic polyimides are the most popular heat-resistant polymers. They are used in various industries, such as electrical insulation, space, aviation, electronics, and industries using gas permeation. Polyimides have attracted much attention as matrix resins for fiber-reinforced composites because of their ability to withstand high-temperature environments.<sup>1</sup> As aerospace technology progresses, high performance materials are needed for many special purpose applications. These materials in many cases need to have good thermoxidative stability, mechanical strength at high temperatures, and good retention of their properties even after a severe aging process. However, most synthetic polymers that fit these requirements are too brittle. Therefore, the question of how best to toughen these polymers becomes very critical. Many aromatic polyimides end

capped with maleimido and nadimido groups have been developed in the last two decades. The bis-maleimides (BMI) prepared from 4,4'-diaminodiphenylmethane is one of them<sup>2,3</sup> and has high  $T_g$ , high modulus, as well as thermal stability, once completely cured. However, this type of BMI is very brittle. Various modified BMIs have been developed<sup>3–10</sup> using different methodologies.

The first attempt to develop nadimide end-capped polyimides was carried out by Lubowitz<sup>11</sup> using the standard poly(amic acid) (PAA) synthesis and marketed by Ciba-Geigy as P13N resin ( $M_n = 1300$ ). However, this type of resin has various shortcomings, such as instability of PAA, poor resin flow, residual solvent, and water evolution, which create porosity in composites. Improved nadimide type resins such as PMR-15, PMR-11, LARC 160, and nadimidized bisaspartimides were later developed.<sup>12–16</sup>

PMR-15<sup>12,13</sup> polymer matrix composites have been used to replace metal components in aircraft engines where operating temperatures do not exceed 290°C. Despite the wide spread use of PMR-15 it

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does not perform well in oxidative environments where temperatures are greater than 290°C. Thus, there is a continuous demand for improved matrix resins for numerous civil and defense applications.

An effective means of increasing the modulus and improving the solvent resistance in linear polymers has been by incorporating carbonyl and ether groups between the aromatic ring in the backbone. Thus poly(aryl ether ketones) (PEKs) are among the high-performance polymers currently being examined for use in aerospace and electronic applications. The research in our laboratory<sup>19</sup> and others<sup>17,18</sup> has focused on producing soluble, thermally crosslinkable PEK polymers or oligomers, which when cured display improved solvent resistance. In this paper we report synthesis and characterization of a homologous series of aromatic polyimides containing hexafluoroisopropylidene and keto and ether groups, thermally crosslinkable by addition reactions. Thermal evaluation of polymers is also presented. The incorporation of hexafluoroisopropylidene (HFIP or 6F) groups into polymer backbone is expected<sup>20</sup> to provide increased  $T_g$  and flame resistance with concomitant decreased water absorption without forfeiture of thermal stability.

## EXPERIMENTAL

### Materials

Aromatic diamine, 1,3-bis(4-aminophenoxy-4'-benzoyl)benzene (1,3-BAPBP) (I), m.p. 162–164°C (Lit.<sup>21</sup>, m.p. 161.5–164°C) was synthesized by us<sup>22</sup> from chlorobenzene and isophthaloyl chloride, followed by reaction of the resulting 1,3-bis(4-chloro-4'-benzoyl)benzene with 4-hydroxyacetanilide and hydrolysis. Maleic anhydride (II<sub>m</sub>) (Aldrich) was purified by recrystallization from chloroform, m.p. 54°C. Endobicyclo[2,2,1]-hept-5-ene-2,3-dicarboxylic anhydride (nadic anhydride) (II<sub>n</sub>) (Aldrich) was purified by recrystallization from acetic anhydride, m.p. 175°C (d). Aromatic dianhydrides were purified by vacuum sublimation or recrystallization from acetic anhydride and tested before use on a differential thermal analyzer (DTA). 4,4'-Hexafluoroisopropylidene diphthalic anhydride (6FDA) (III) showed melting at 239–240°C. 3,3',4,4'-Tetracarboxylic benzophenone dianhydride (BTDA) (IV) and pyromellitic dianhydride (V) showed a single endotherm at 223°C and 285–287°C, respectively. The solvent *N,N'*-dimethylacetamide (DMAC) was dried over phosphorus pentoxide and distilled under vacuum. Chloroform was dried by distillation over phosphorus pentoxide, b.p. 61°C.

### Techniques

Melting points were uncorrected and determined in a capillary tube by using an optical microscope melting point apparatus. Infrared (IR) spectra were recorded on a Shimadzu spectrometer IR-435 on a KBr disk. Elemental analysis was performed on a Heraeus CHN elemental analyzer. Dynamic thermogravimetric analysis (TGA) and differential thermal analysis (DTA) were performed on a Perkin-Elmer System 7/4 thermal analysis instrument attached to a Perkin-Elmer computer data station, heating rate 20°C/min; gas flow 100 mL/min; and sample weight 3–5 mg. Proton magnetic resonance (<sup>1</sup>H-NMR) spectra were recorded on a Perkin-Elmer R32 (90 MHz) spectrometer. The chemical shift ( $\delta$ ) is given in ppm with tetramethylsilane (TMS) as an internal standard.

### General Synthesis

The reactions were carried out in a three-necked R.B. flask equipped with a nitrogen inlet, a magnetic stirrer, a teflon-coated bar, and a calcium chloride guard tube. The reagents were added in the sequences and stoichiometrics indicated below and in Table I. The method described is that which gave optimum yield of the title compound and is not the only method by which these compounds can be produced. As an example of the general method, the preparation of bismaleamic acid (VI), and its *in situ* imidization to bismaleimide (XII), and thermal polymerization is given.

Into a continuously stirred solution of 1,3-bis(4-aminophenoxy-4'-benzoyl)benzene (I) (1 g, 0.002 mol) in DMAC (7.5 mL), dry nitrogen gas was bubbled. To this solution, granular 4,4'-hexafluoroisopropylidene diphthalic anhydride (0.444 g, 0.001 mol) (6FDA) (III) was added and the solution was stirred for 0.5 h. To the yellow solution, maleic anhydride (II<sub>m</sub>) (0.206 g, 0.0021 moles) was added and the stirring continued for 1 h to give the bismaleamic acid (VI).

ANAL: Calculated for C<sub>90</sub>H<sub>58</sub>N<sub>4</sub>O<sub>18</sub>F<sub>6</sub>: C, 67.67%; H, 3.63%; N, 3.51%. Found: C, 67.49%; H, 3.58%; N, 3.60%.

### *In Situ* Imidization and Thermal Polymerization

The clear yellow solution of bismaleamic acid (VI) in DMAC was taken in an aluminum cup and placed in a preheated air oven maintained at 65–70°C and the solvent was allowed to evaporate off. The cyclodehydration of the bismaleamic acid (VI) was per-

**Table I** Preparation and Elemental Analysis of Bismaleamic Acids and Bisnadic Acids

Sample	Amount of Reactants									Elemental Analysis		
	Diamine (I)		Dianhydride		End-Cap			Molecular Formula				
	Mol	g	Type	Mol	g	Type	Mol		g	C (%)	H (%)	N (%)
VII	0.002	1	6FD	0.001	0.444	NA	0.0021	0.344	C <sub>102</sub> H <sub>70</sub> N <sub>4</sub> F <sub>6</sub> O <sub>20</sub>	Calcd.: 68.61 Found: 69.02	3.92 3.88	3.14 3.25
IX	0.002	1	BTDA	0.001	0.322	NA	0.0021	0.344	C <sub>99</sub> H <sub>70</sub> N <sub>4</sub> O <sub>21</sub>	Calcd.: 72.00 Found: 71.92	4.25 4.30	3.39 3.35
VIII	0.002	1	BTDA	0.001	0.322	MA	0.0021	0.205	C <sub>89</sub> H <sub>58</sub> N <sub>4</sub> O <sub>21</sub>	Calcd.: 70.36 Found: 70.52	3.82 3.76	3.69 3.75
XI	0.002	1	PMDA	0.001	0.218	NA	0.0021	0.344	C <sub>92</sub> H <sub>66</sub> H <sub>4</sub> O <sub>20</sub>	Calcd.: 71.41 Found: 71.51	4.27 4.32	3.62 3.58
X	0.002	1	PMDA	0.001	0.218	MA	0.0021	0.205	C <sub>82</sub> H <sub>54</sub> N <sub>4</sub> O <sub>20</sub>	Calcd.: 69.59 Found: 69.92	3.82 3.88	3.96 4.05

formed at 160°C for 0.5 h to give bismaleimide (XII). Thermal polymerization of XII was carried out at 225°C for 1.5 h and at 290°C for 0.5 h to give a tough, brown polymer (XVIII).

By a similar method, bismaleamic acid (VIII) and (X) were synthesized by the reaction of 1,3-bis(4-aminophenoxy-4'-benzoyl)benzene (I) with BTDA (IV) and maleic anhydride (MA) (IIm), and with PMDA (V) and MA (IIm), respectively. The bismaleamic acids were cyclodehydrated *in situ* to bismaleimides (XIV) and (XVI), and thermally polymerized to polymer (XIX) and (XX), respectively.

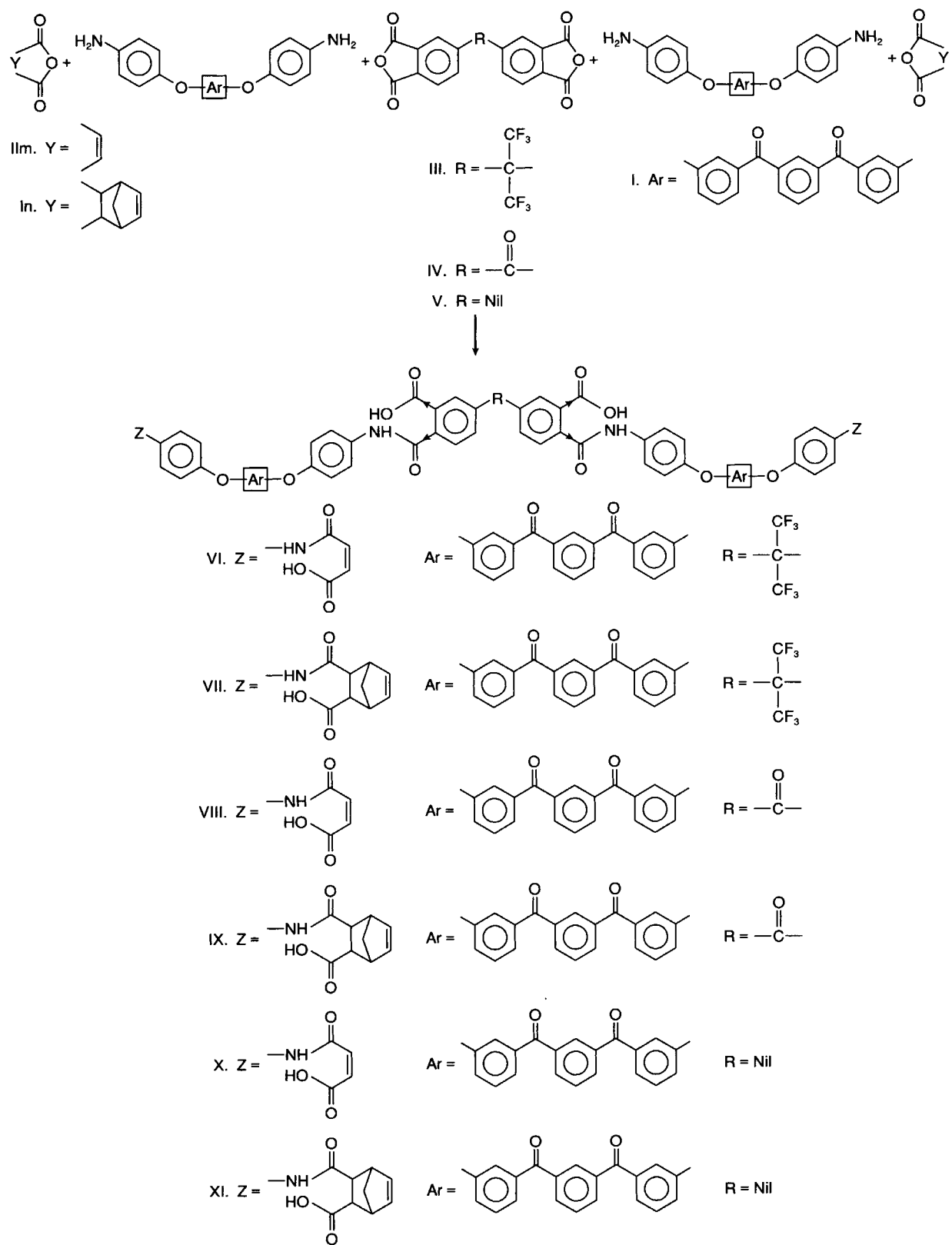
Using a similar method, various bisnadic acids (VII), (IX), and (XI) were prepared from the diamine (I), aromatic bisanhydrides, and nadic anhydride (IIn), respectively. The details of the preparation are given in Table I.

## RESULTS AND DISCUSSION

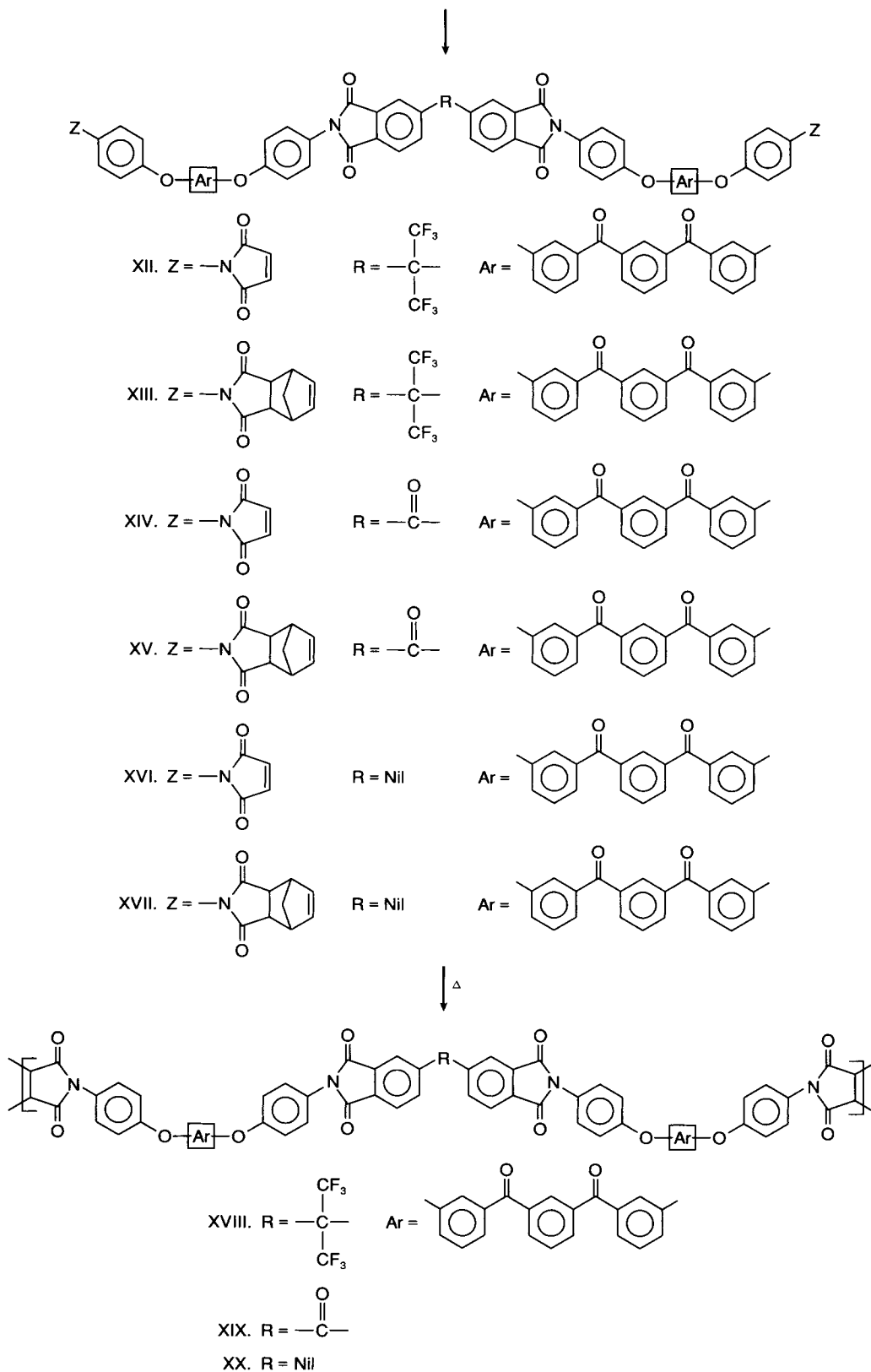
Aromatic diamine containing keto and ether groups, viz., 1,3-bis(4-aminophenoxy-4'-benzoyl)benzene (1,3-BAPBP) (I) was synthesized by our modified method<sup>22</sup> and characterized by FT-IR, Mass, and <sup>1</sup>H-NMR spectroscopy.

Schemes 1 and 2 outline the synthesis of polymers (XVII), (XIX), (XX), (XXI), (XXII), and (XXIII) by involving the reaction of diamine (1,3-BAPBP) (I) with aromatic dianhydrides (III), (IV), and (V) and end capping with maleic anhydride (IIm) or nadic anhydride (IIn), respectively, followed by *in situ* cyclodehydration and thermal polymerization.

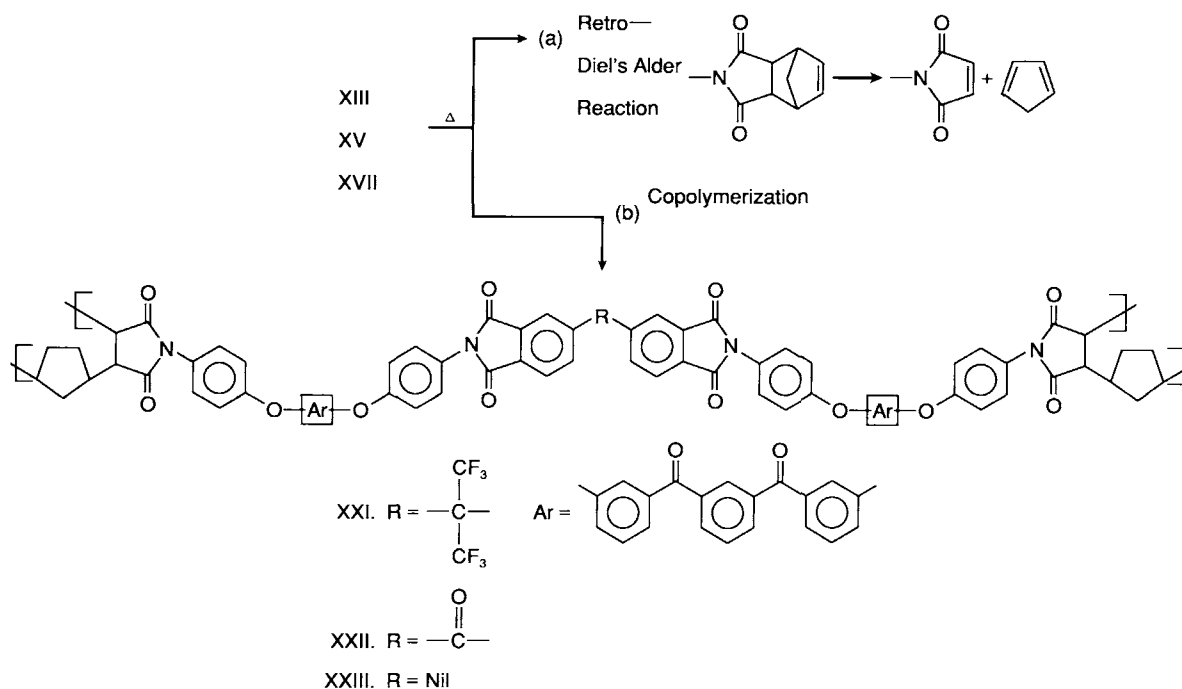
1,3-Bis(4-aminophenoxy-4'-benzoyl)benzene (1,3-BAPBP) (I) was reacted in DMAC with a calculated amount of hexafluoroisopropylidene diphthalic anhydride (6FDA) (III) and maleic anhydride (IIm) to give the corresponding bridged bismaleamic acid (VI). The bridged bismaleamic acid (VIII) and (X) were similarly synthesized from BTDA (IV) and PMDA (V), respectively. All the synthesized maleamic acids were characterized by elemental analysis (Table I), <sup>1</sup>H-NMR, and IR spectroscopy. A <sup>1</sup>H-NMR spectrum of bismaleamic acid (VI) showed the absence of amino protons of the diamine (I) and presence of a quartet (four protons) at 6.50–6.08 ppm corresponding to the *cis* olefinic protons. Also observed were two multiplets at 7.10–6.90 ppm, and 7.50–7.45 ppm due to the aromatic protons of diamino and 6FDA moieties. The integration matched the expected spectra. This confirmed the attachment of the two maleic anhydride units at the amino terminals to give the bismaleamic acid (VI). The infrared spectrum of bismaleamic acid (VI) showed the presence of carboxylic and olefinic groups, as can be inferred from the bands observed at 3400–3150 cm<sup>-1</sup>, 1715 cm<sup>-1</sup>, and a band at 1584 cm<sup>-1</sup>, respectively. The *in situ* thermal cyclodehydration of the bismaleamic acid (VI) at 160°C gave the bridged bismaleimide (XII). The infrared spectrum of the bismaleimide (XII) showed [Fig. 1(D)] the characteristic<sup>16</sup> imide carbonyl bands at 1782, 1716, and 742 cm<sup>-1</sup>. Also presence of keto (1652 cm<sup>-1</sup>), ether (1242 cm<sup>-1</sup>) and the aromatic groups (1498 cm<sup>-1</sup>) were clearly observed. Figure 1(C) and (E) similarly showed the characteristic IR-spectra of bridged bismaleimide (XVI) and (XIV), respectively.



Scheme 1



**Scheme 1** (continued from the previous page)



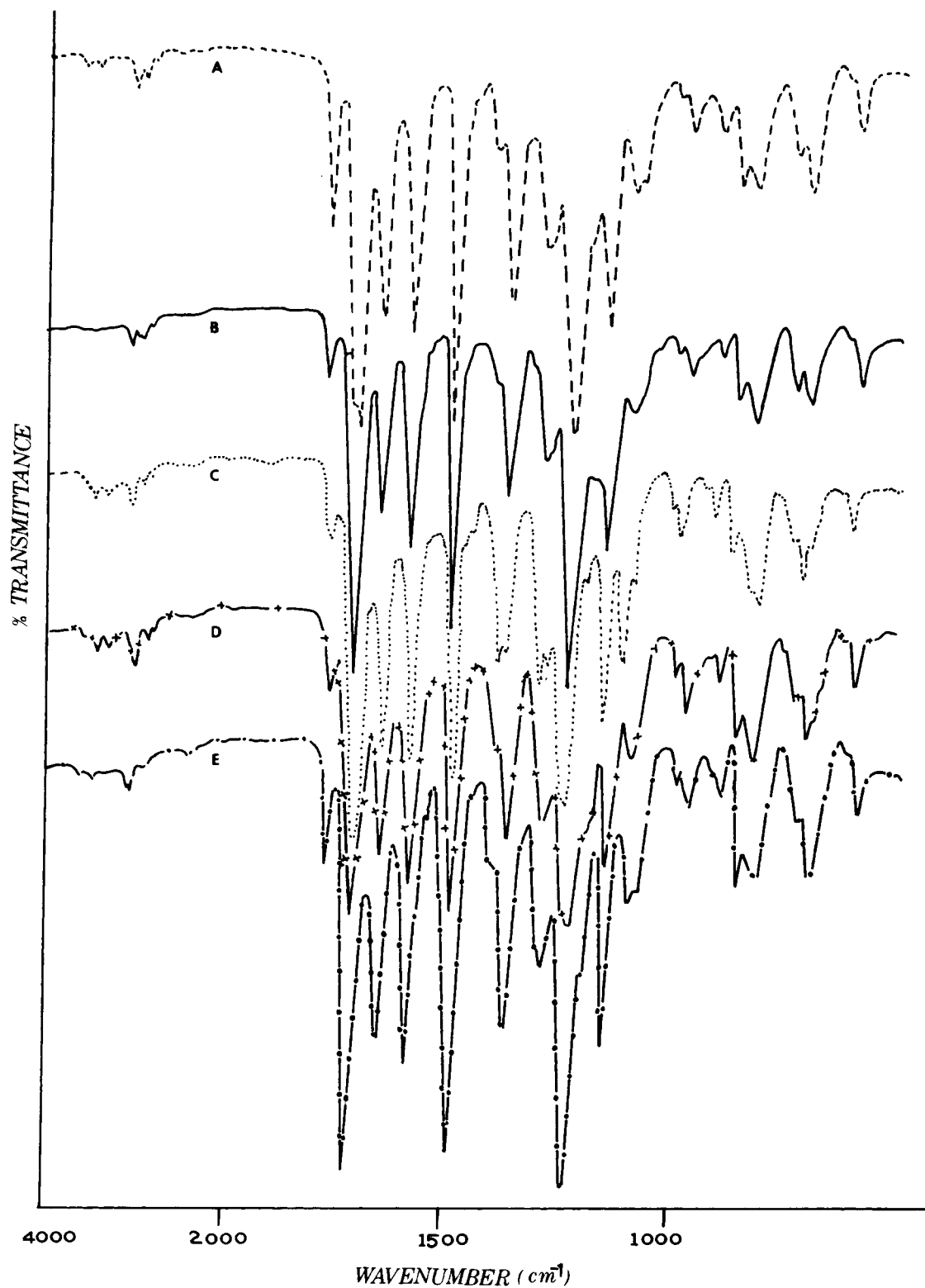
Scheme 2

1,3-Bis(4-aminophenoxy-4'-benzoyl)benzene (1,3-BAPBP) (I) was reacted in DMAC with a calculated amount of pyromellitic dianhydride (V) and nadic anhydride (II<sub>n</sub>) to give the corresponding bridged bisnadidic acid (XI). The bridged bisnadidic acid (VII) and (IX) were similarly synthesized from 6FDA (III) and BTDA (IV), respectively. All the synthesized nadic acids were characterized by elemental analysis (Table I), <sup>1</sup>H-NMR, and IR spectroscopy. A <sup>1</sup>H-NMR spectrum of bisnadidic acid (XI) showed the absence of amino protons singlet [observed in diamine (I) at 4.82 ppm and exchangeable to D<sub>2</sub>O] and presence of two multiplets at 6.17–6.00 ppm corresponding to the four olefinic protons, the eight aliphatic protons at 3.35 and 3.10–3.00 ppm, and a four protons multiplet at 1.50 ppm of the norbornene bridge-head methylene. These, together with signals at 10.45 and 9.75 ppm, due to carboxylic and amido protons confirmed the nadic anhydride attachment at the amino terminals. Besides these signals, the rest of the <sup>1</sup>H-NMR spectrum of (XI) showed the expected integration of the aromatic protons. The infrared spectrum of bisnadidic acid (XI) showed [Fig. 2(A)] the presence of carboxylic and olefinic groups, as can be inferred from the bands observed at 3450–3150 cm<sup>-1</sup>, 1712 cm<sup>-1</sup>, and a band at 1584 cm<sup>-1</sup>, respectively. The *in situ* thermal cyclodehydration of bisnadidic acid (XI) at 160°C gave the bridged bisnadimide (XVII). The infrared spectrum of the bis-

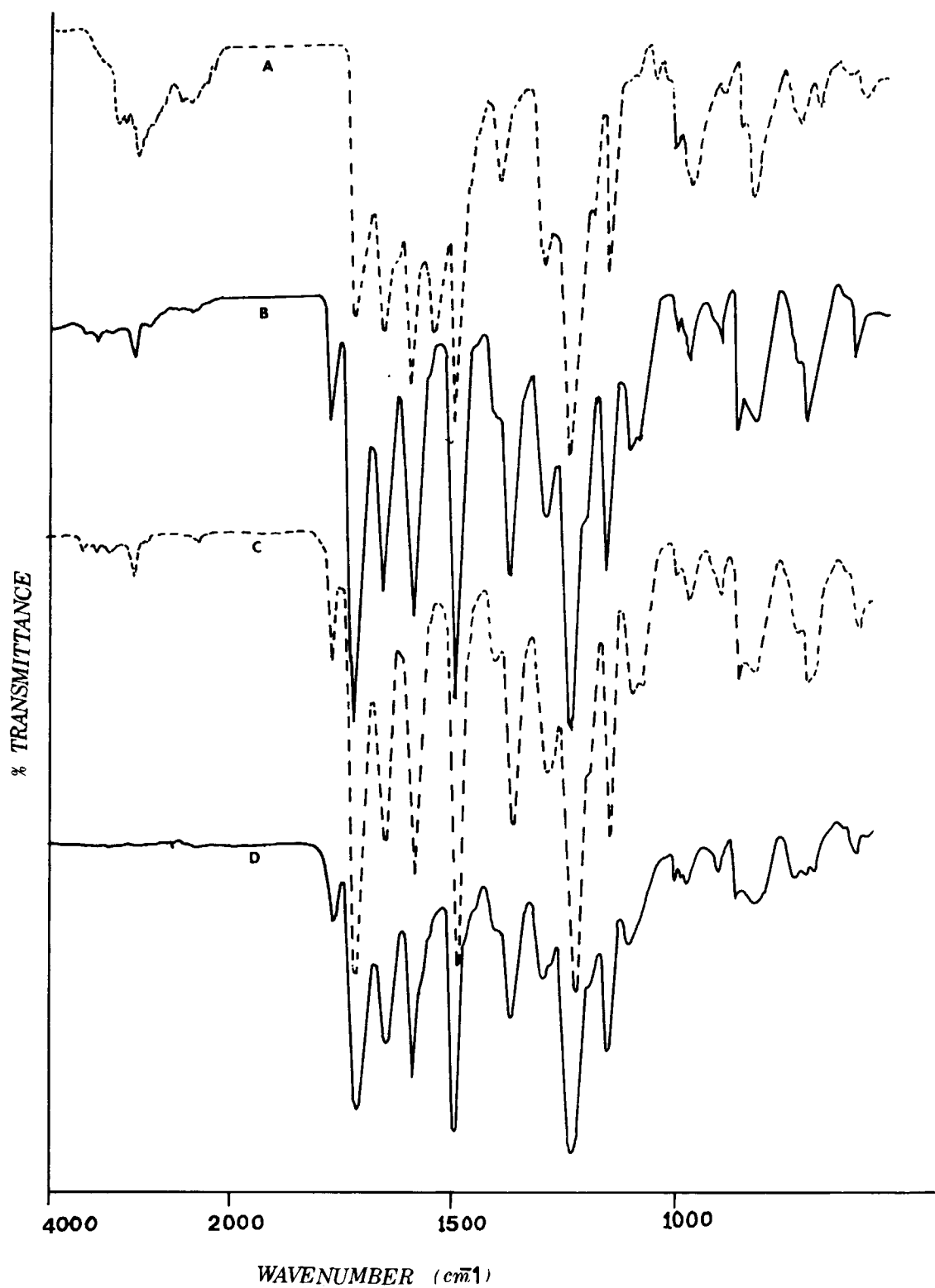
nadimide (XVII) showed [Fig. 2(B)] the characteristic imide carbonyl bands at 1780, 1718, and 742 cm<sup>-1</sup>. The presence of keto (1654 cm<sup>-1</sup>), ether (1245 cm<sup>-1</sup>), and aromatic groups (1500 cm<sup>-1</sup>) were clearly observed. Figure 1(A) and (B) similarly showed the characteristic IR-spectra of bridged bisnadimide (XV) and (XIII), respectively.

### Thermal Polymerization

Thermal polymerization of bismaleimides (XII), (XIV), and (XVI) and bisnadimides (XIII), (XV), and (XVII) was performed above 225°C to form tough, crosslinked polymers. The curing cycles were selected based on our earlier studies.<sup>16</sup> The bismaleimide (XII) was *in situ* thermally polymerized in an air oven maintained at 225°C for 1.5 h. The temperature of the oven was then raised for further curing to 290°C for 0.5 h. A tough, brown polymer (XVIII) was obtained. Similarly, *in situ* thermal polymerization of bismaleimide (XIV) and (XVI) and bisnadimide (XIII), (XV), and (XVII) was carried out thermally in an air oven. Tough polymers (XIX), (XX), (XXI), (XXII), and (XXIII), respectively, were obtained. The thermal polymerization was monitored using IR spectroscopy on a KBr disk heated along with the sample. As a representative example, Figure 2(C) and (D) shows the pat-

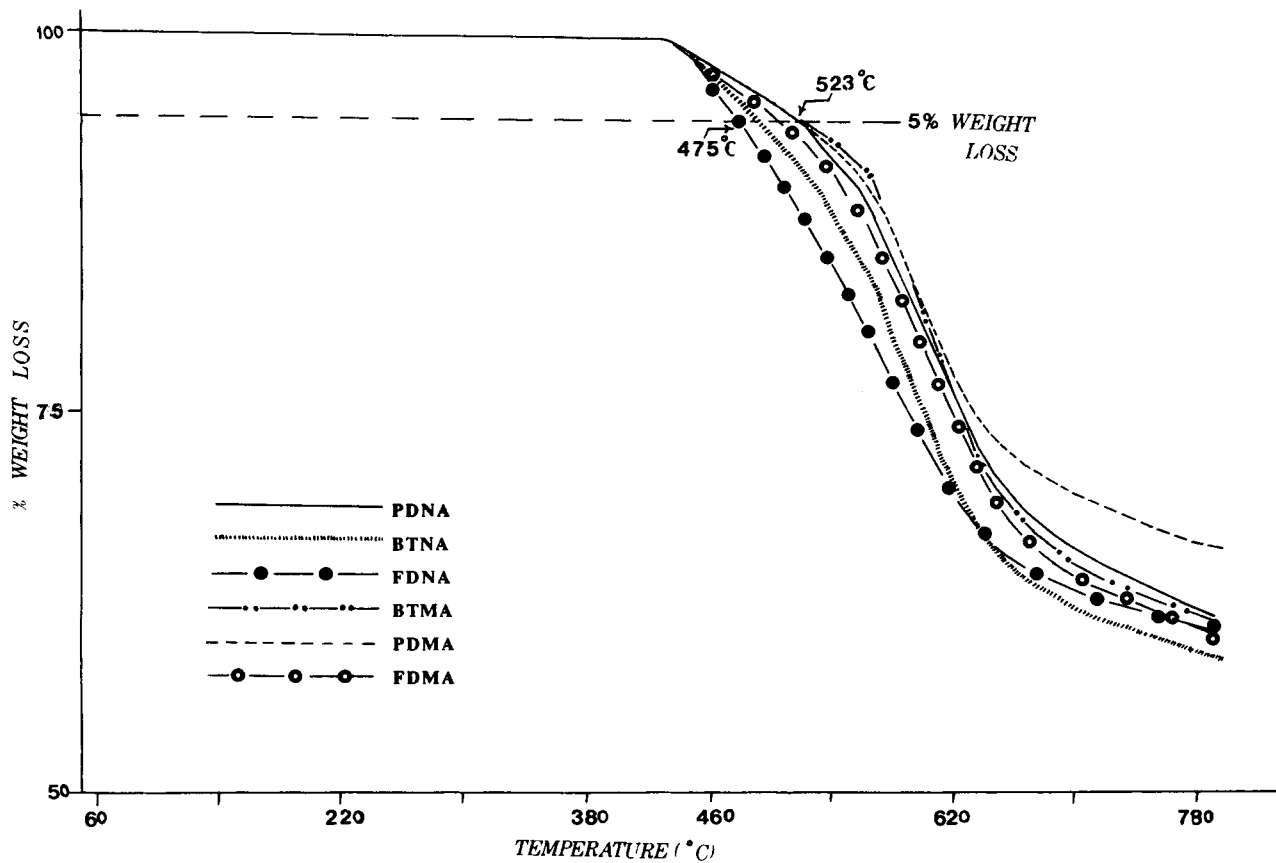


**Figure 1** Infrared spectrum of bridged bismaleimides and bisnadimides on a KBr disk: (A) (XV) (---); (B) (XIII) (—); (C) (XVI) (···); (D) (XII) (-+--); (E) (XIV) (-·-·).

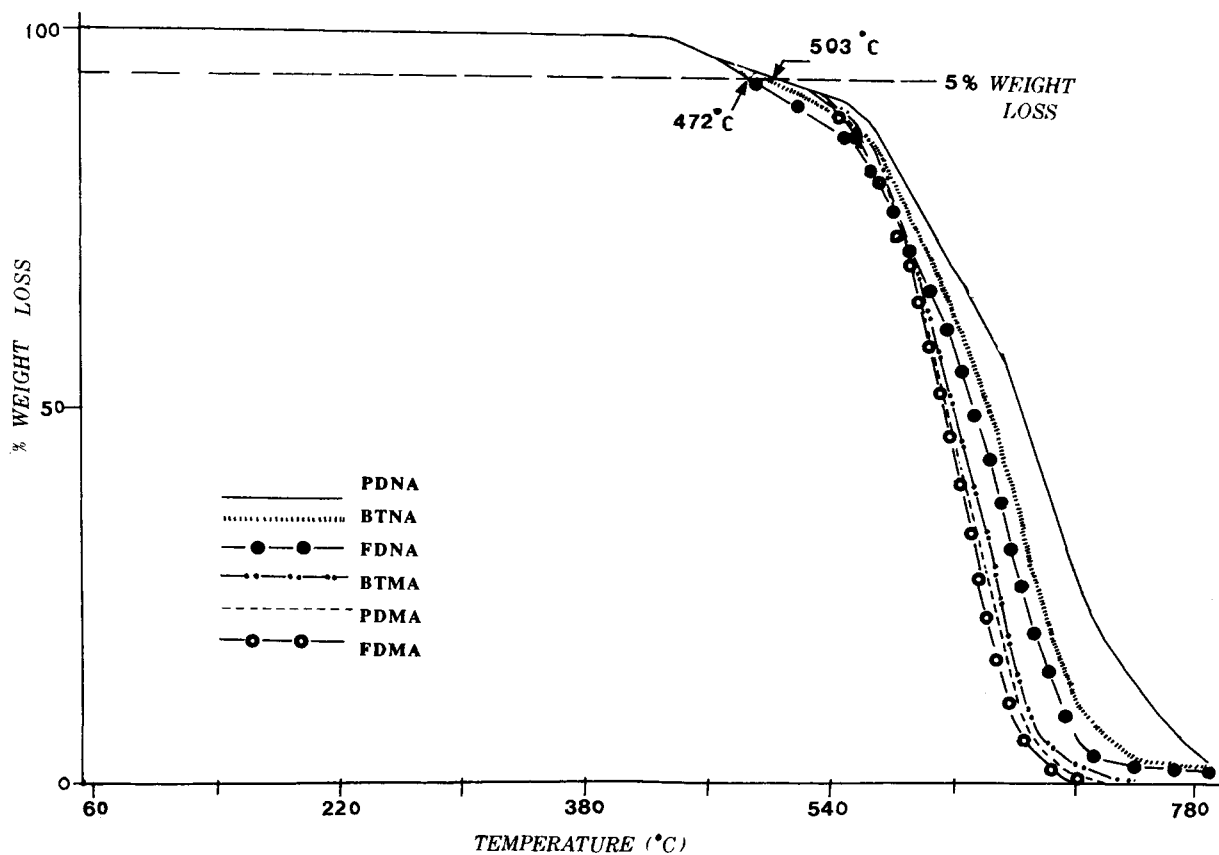


**Figure 2** Infrared spectrum on a KBr disk of: (A) bridged bisnadinic acid (XI); (B) bridged bisnadimide (XVII); (C) after thermal polymerization of XVII at 225°C for 1.5 h; (D) polymer XXIII.





**Figure 3** Dynamic thermogravimetric analysis in nitrogen of polymers: XVIII (FDMA); XIX (BTMA); XX (PDMA); XXI (FDNA); XXII (BTNA); and XXIII (PDNA).



**Figure 4** Dynamic thermogravimetric analysis in air of polymers: XVIII (FDMA); XIX (BTMA); XX (PDMA); XXI (FDNA); XXII (BTNA); and XXIII (PDNA).

tern of IR obtained at 225°C for 1.5 h and at 290°C for 0.5 h. A general broadening of the IR bands at this stage indicated polymerization. It was suggested earlier that thermal crosslinking for nadimide end caps proceed partially<sup>12a</sup> or completely<sup>23</sup> via an initial retro Diels–Alder dissociation (at and above 275°C). Therefore the polymers formed from bisnadimides have saturated crosslinking units, obtained by partial or complete copolymerization of *in situ* formed maleimide and cyclopentadiene moieties (Scheme 2).

The synthesized polymers were evaluated for their thermal stability using dynamic thermogravimetric analysis (TGA) in nitrogen and air atmosphere. The respective thermograms are shown in Figures 3 and 4. In Table II, polymer decomposition temperature (PDT), temperature of maximum weight of rate loss (PDT<sub>max</sub>, obtained from the differential thermograms recorded simultaneously), and anaerobic char yield at 800°C are given. These polymers showed PDT at 430–435°C and PDT<sub>max</sub> at 575–600°C. The anaerobic char yield of the polymers was in the range of 60–67% at 800°C. In air (Fig. 4) all the polymers decompose above 620°C without any char or negligible char yield at 800°C. The rate of decomposition, however, was slower than previously reported<sup>19</sup> bismaleimides and bisnadimides containing keto and ether groups. It may be pointed out here that all the polymers showed a weight loss (7–9%) between 430–540°C due to the aliphatic nature of the crosslinking units. The pattern of decomposition observed above 540°C was similar to aromatic polyimides. This study demonstrated that in air (Fig. 4) polymers (XXI) (FDNA), (XXII) (BTNA), and (XXIII) (PDNA)

obtained from the nadimide end caps showed decomposition at higher temperatures (Table II) than the respective polymers (XVIII) (FDMA), (XIX) (BTMA), and (XX) (PDMA) obtained from maleimide end caps. The observed order of thermal stability of the synthesized polymers was PDNA > BTNA > FDNA > PDMA > BTMA > FDMA, depicting the higher thermal stability of pyromellitic dianhydride bridged units in each category. In nitrogen, however, the polymers obtained from maleimide end caps, in general, showed higher thermal stability. The order of their thermal stability was also similar, irrespective of the nature of the end caps, that is, PDMA > PDNA // BTMA > BTNA // FDMA > FDNA. The observed improved thermooxidative stability for polymers derived from nadimide end caps may be explained by the fact that during decomposition, the crosslinked units change in such a way that the weight loss is comparatively less, which may be attributed to particular aromatization reactions and to the presence of di-keto and ether groups. The bridging unit of the used dianhydride also plays an important role, that is, in PMDA where the bridged diimide groups are separated without any linking group, a higher thermal stability was observed. In the other bridged diimides, the keto linking group from BTDA showed improved thermal stability than the  $-\text{C}(\text{CF}_3)_2-$  group of 6FDA.

All the synthesized polymers were found insoluble in DMAC, DMSO, and chloroform. The developed polymers are potential candidates for the preparation of fiber-reinforced composites and materials for high-temperature applications. It is anticipated that using the similar methodology, PMR-type matrix

**Table II** Dynamic Thermogravimetric Analysis of Polyimides in Nitrogen and Air

Polymer (Designation*)	Aromatic Dian- hydride	End Cap	In Nitrogen				In Air			
			PDT (°C)	PDT <sub>5</sub> <sup>b</sup> (°C)	PDT <sub>max</sub> (°C), W <sub>r</sub> <sup>a</sup>	Char Yield		PDT (°C)	PDT <sub>5</sub> (°C)	
						700°C	800°C			
XVIII (FDMA)	6FA	MA	430	503	600 (80)	60.5	64	430	503	
XXI (FDNA)	6FA	NADA	430	475	575 (79)	61	64	430	472	
XIX (BTMA)	BTDA	MA	435	523	605 (79)	62	65	435	503	
XXII (BTNA)	BTDA	NADA	435	495	580 (78)	60	62	435	495	
XX (PDMA)	PMDA	MA	435	525	600 (82)	67	71	435	503	
XXIII (PDNA)	PMDA	NADA	435	525	600 (79)	63	66	435	503	

PD, pyromellitic dianhydride (PMDA); BT, benzophenone tetracarboxylic dianhydride (BTDA); FD, hexafluoroisopropylidene dianhydride (6FDA); MA, maleic anhydride; and NA, nadic anhydride.

<sup>a</sup> W<sub>r</sub> = Percentage weight remaining at that temperature.

<sup>b</sup> PDT<sub>5</sub> = Temperature at which 5% weight loss occurs.

resins can be made that will be useful for aircraft engines.

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