

Synthesis and Evaluation of Novel Polyimides Derived from Spirobichroman Diether Anhydride

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ABSTRACT: A spirobichroman structure-containing diether anhydride (SBCDA), 6,6'-bis(3,4-dicarboxyphenoxy)-4,4,4',4',7,7'-hexamethyl-2,2'-spirobichroman dianhydride, was prepared by the nucleophilic nitrodisplacement of 4-nitrophthalonitrile with the phenolate ion of 6,6'-dihydroxy-4,4,4',4',7,7'-hexamethyl-2,2'-spirobichroman, followed by alkaline hydrolysis of the intermediate tetranitrile and dehydration of the resulting tetraacid. A series of high molecular weight poly(ether imide)s with inherent viscosities between 0.45 and 1.28 dL/g were synthesized from SBCDA and various aromatic diamines via a conventional two-stage procedure that included ring-opening polyaddition in *N,N*-dimethylacetamide (DMAc) to give poly(amic acid)s, followed by thermal cyclization to poly(ether imide)s. The intermediate poly(amic acid)s had inherent viscosities of 0.70–1.50 dL/g. Except for the poly(ether imide) obtained from *p*-phenylenediamine, the other poly(ether imide)s were soluble in various organic solvents and could be solution-cast into transparent, flexible, and tough films. These poly(ether imide)s had glass transition temperatures in the range 175–262°C and showed no significant decomposition below 420°C, with 10% weight loss being recorded above 446°C in nitrogen or air. © 1997 John Wiley & Sons, Inc. *J Polym Sci A: Polym Chem* **35**: 2801–2809, 1997

Keywords: spirobichroman; diether anhydride; poly(ether imide)s

INTRODUCTION

Aromatic polyimides are known for their excellent thermal stability and unique combination of mechanical and electronic properties.¹ They have been applied extensively in the electronic and aerospace industries. The imide heterocyclic unit and the aromatic residue impart rigidity to the polymer backbone and also afford strong intermolecular interactions. This usually makes aromatic polyimides insoluble and infusible. Therefore, it is difficult to fabricate them in the fully imidized

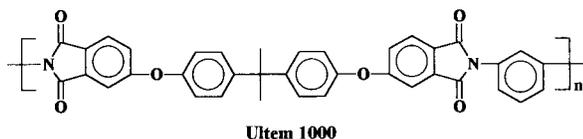
form. In fact, they are usually processed in the form of their poly(amic acid) precursors followed by chemical or thermal imidization. Problems can arise because the poly(amic acid)s are thermally and hydrolytically unstable. The water byproduct from imidization can also form microvoids in the final product. Therefore, considerable research has been undertaken in order to identify new ways to circumvent these restrictions.

The introduction of flexible group and/or bulky units in the polymer backbone has been a general approach to change the chemical structure of polyimides.^{2–10} Poly(ether imide)s were developed as a result of research interests in aromatic nucleophilic displacement chemistry combined with a perceived marketplace need for high performance polymers which could be readily fabricated by standard plastics extrusion and injection molding

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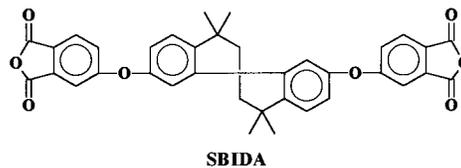
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processes. An important example is Ultem 1000 developed and commercialized by General Electric Co.,¹¹ which exhibits reasonable thermal stability and good mechanical properties together with good moldability.



Research for new polyimides with improved processability and higher T_g s than that of Ultem 1000 has been a continuing effect since the commercialization of Ultem 1000. Cella and Falser¹² have reported a new dianhydride, spirobis(indane) diether anhydride (SBIDA). The polyimide prepared from SBIDA and an aromatic diamine had a higher T_g than that made from bisphenol A diether anhydride and the same diamine. It is conceivable that the structural rigidity of SBIDA contributes to the increase in the T_g of those polyimides. However, as demonstrated in a more detailed report by Chao and Barren,¹³ polyimides derived from SBIDA and the diamines of various size and shape invariably contained cyclic oligomers which usually made their compression-molded films not have suitable ductility. The formation of cyclic contaminants should be related to the rigidity of the spirobiindane structure. The present article reports a new dianhydride (SBCDA) having the structure **4** as shown in Scheme 1 and new polyimides derived from it. The dianhydride SBCDA was synthesized from 6,6'-dihydroxy-4,4,4',4',7,7'-hexamethyl-2,2'-spirobichroman (**1**) and 4-nitrophthalonitrile as starting material according to a literature procedure.¹⁴ The methyl-substituted spirobichroman unit containing diol **1** can be readily obtained from the cyclocondensation of methylhydroquinone with acetone in the presence of an acid catalyst,¹⁵⁻¹⁸ and now is commercially available from a local supplier. This dianhydride, SBCDA, has a spirobichroman structure in the center of the molecule. It should have a higher conformational mobility than SBIDA due to the six-membered heterocycle. Furthermore, the molecular model of SBI shows it makes a sharp kink in the repeat unit that enables two end groups to come close to each other. In comparison, the 6,6-dihydroxyspirobichroman system shows a larger dihedral angle ($\text{HO} \cdots \text{C}_{\text{spiro}} \cdots \text{OH}$), making oligomer ring closure more

difficult. Therefore, the polyimides derived from SBCDA have few cyclic oligomers and exhibit many attractive properties. This report summarizes our findings.



EXPERIMENTAL

Materials

6,6'-Dihydroxy-4,4,4',4',7,7'-hexamethyl-2,2'-spirobichroman (HMSBC-diol; **1**) (Tokyo Chemical Industry Co.; TCI), 4-nitrophthalonitrile (TCI), and dimethyl sulfoxide (DMSO; Aldrich) were used as supplied. *N,N*-Dimethylacetamide (DMAc) was purified by distillation under reduced pressure in the presence of calcium hydride and stored over 4 Å molecular sieves.

p-Phenylenediamine (**5a**; TCI) and benzidine (**5c**; TCI) were purified by sublimation. *m*-Phenylenediamine (**5b**; Janssen) was vacuum-distilled prior to use. 4,4'-Oxydianiline (**5d**; TCI), 3,4'-oxydianiline (**5e**; TCI), 4,4'-methylenedianiline (**5f**; TCI), and 1,4-bis(4-aminophenoxy)benzene (**5g**) were used as received.

According to a reported method,¹⁹ 4,4'-bis(4-aminophenoxy)biphenyl (**5h**), 2,2-bis[4-(4-aminophenoxy)phenyl]propane (**5i**), and bis[4-(4-aminophenoxy)phenyl]sulfone (**5j**) were prepared by aromatic nucleophilic substitution of *p*-chloronitrobenzene with the bisphenoxide ion of 4,4'-biphenol, 2,2-bis(4-hydroxyphenyl)propane, and bis(4-hydroxyphenyl)sulfone, respectively, followed by reduction of the intermediate dinitro compounds.

Monomer Synthesis

6,6'-Bis(3,4-dicyanophenoxy)-4,4,4',4',7,7'-hexamethyl-2,2'-spirobichroman (**2**)

In a 500-mL round-bottomed flask, 15 g (0.04 mol) of HMSBC-diol (**1**) and 15 g (0.08 mol) of 4-nitrophthalonitrile (15 g; 0.08 mol) were dissolved in 230 mL of DMSO. Anhydrous potassium carbonate (11.5 g; 0.08 mol) was added, and then the suspension was stirred at room temperature for

48 h. The reaction mixture was then poured into 500 mL of deionized water to give a pale-yellow solid product which was washed repeatedly with water and methanol, filtered out, and dried. The product **2** was obtained in the form of a pale-yellow powder in 82% yield (21 g); mp 263–265°C. IR (KBr) [Fig. 1(A)]: 2230 (C≡N str.), 1230, 1160 cm⁻¹ (C—O str.).

6,6'-Bis(3,4-dicarboxyphenoxy)-4,4,4',4',7,7'-hexamethyl-2,2'-spirobichroman (3)

In a 500-mL round-bottomed flask 16 g (0.026 mol) of tetranitrile **2** was suspended in a solution of 29 g (0.52 mol) potassium hydroxide in 80 mL/80 mL of water–ethanol. Within 2–4 h the solid tetranitrile dissolved. Reflux was continued until the evolution of ammonia had ceased. After filtration, the cooled filtrate was acidified by concentrated HCl to pH = 2–3. The solid tetraacid (**3**) was filtered off and washed thoroughly with deionized water until the filtrate was neutral. Then the product was dried. Yield: 16.7 g (93%). IR (KBr) [Fig. 1(B)]: 2500–3600 (O—H str.), 1711 (C=O str.), 1220, 1158 cm⁻¹ (C—O str.).

6,6'-Bis(3,4-dicarboxyphenoxy)-4,4,4',4',7,7'-hexamethyl-2,2'-spirobichroman Dianhydride (4)

In a 500-mL round-bottomed flask, 16.1 g (0.027 mol) of tetraacid **3** was suspended in 100 mL of glacial acetic acid and 200 mL of acetic anhydride. The mixture was boiled under reflux for 2 h. Then, the mixture was filtered and left to crystallize overnight. The precipitated product (white needles) was filtered out, washed with dry toluene, and dried, to give 11.9 g (78.6%) of dianhydride **4**; mp 285–286°C. IR (KBr) [Fig. 1(C)]: 1852 (asym. C=O str.), 1781 (sym. C=O str.), 1286, 1154 cm⁻¹ (C—O str.). Anal. Calcd for C₃₉H₃₂O₁₀ (660.67): C, 70.90%; H, 4.88%. Found: C, 70.19%; H, 4.95%.

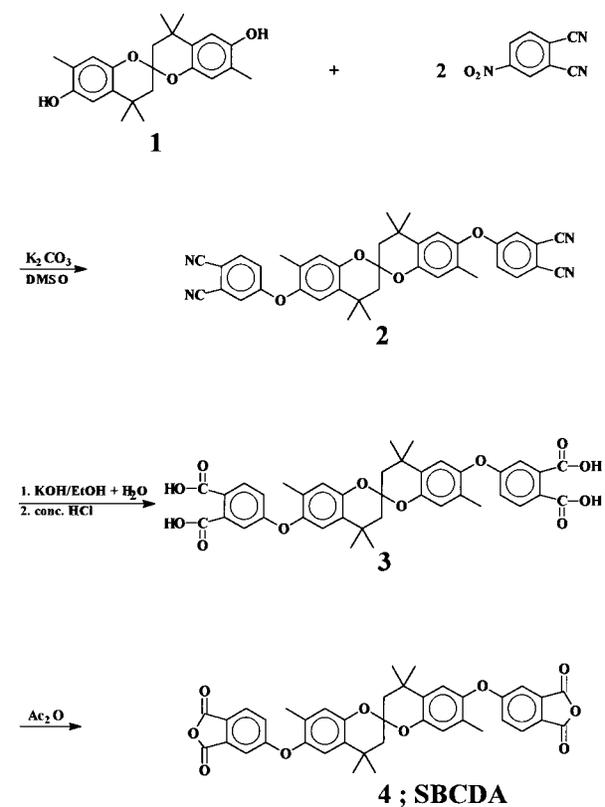
Polymerization

A typical example of polymerization was as follows. The poly(amic acid) was prepared at a concentration of 10% (w/w) solids by the slow addition of dianhydride **4** (0.7674 g; 1.16 mmol) to a stirred DMAc (9.5 mL, about 9 g) solution of 4,4'-oxydianiline (**5d**, 0.2326; 1.16 mmol) at room temperature. After 2 h, the resulting viscous poly(amic acid) solution was characterized with an inherent viscosity of 0.94 dL/g (measured on 0.5

g/dL in DMAc at 30°C). The solution was poured onto a smooth and clean glass dish, which was placed in a 90°C oven for 12 h. The semidried poly(amic acid) film was further dried and imidized by a sequential heating process: 120°C/20 min, 150°C/30 min, 200°C/30 min, and 250°C/30 min. By being soaked in hot water, the polyimide film was lifted off from the glass substrate.

Measurements

Elemental analyses were performed using a Perkin-Elmer Model 2400 C, H, N elemental analyzer. IR spectra were recorded on a Jasco FT/IR-7000 Fourier-transform infrared spectrometer. The inherent viscosities were measured with either an Ubbelohde or a Cannon-Fenske viscometer thermostated at 30°C. The differential scanning calorimetry (DSC) was performed on a Sinku Riko DSC-7000 coupled to a Sinku Riko TA-7000 thermal analyzer in flowing nitrogen (30 cm³/min) at a heating rate of 10°C/min. Thermogravimetry (TG) was conducted with a Rigaku Thermoflex TG-8110 coupled to a Rigaku TAS-100



Scheme 1.

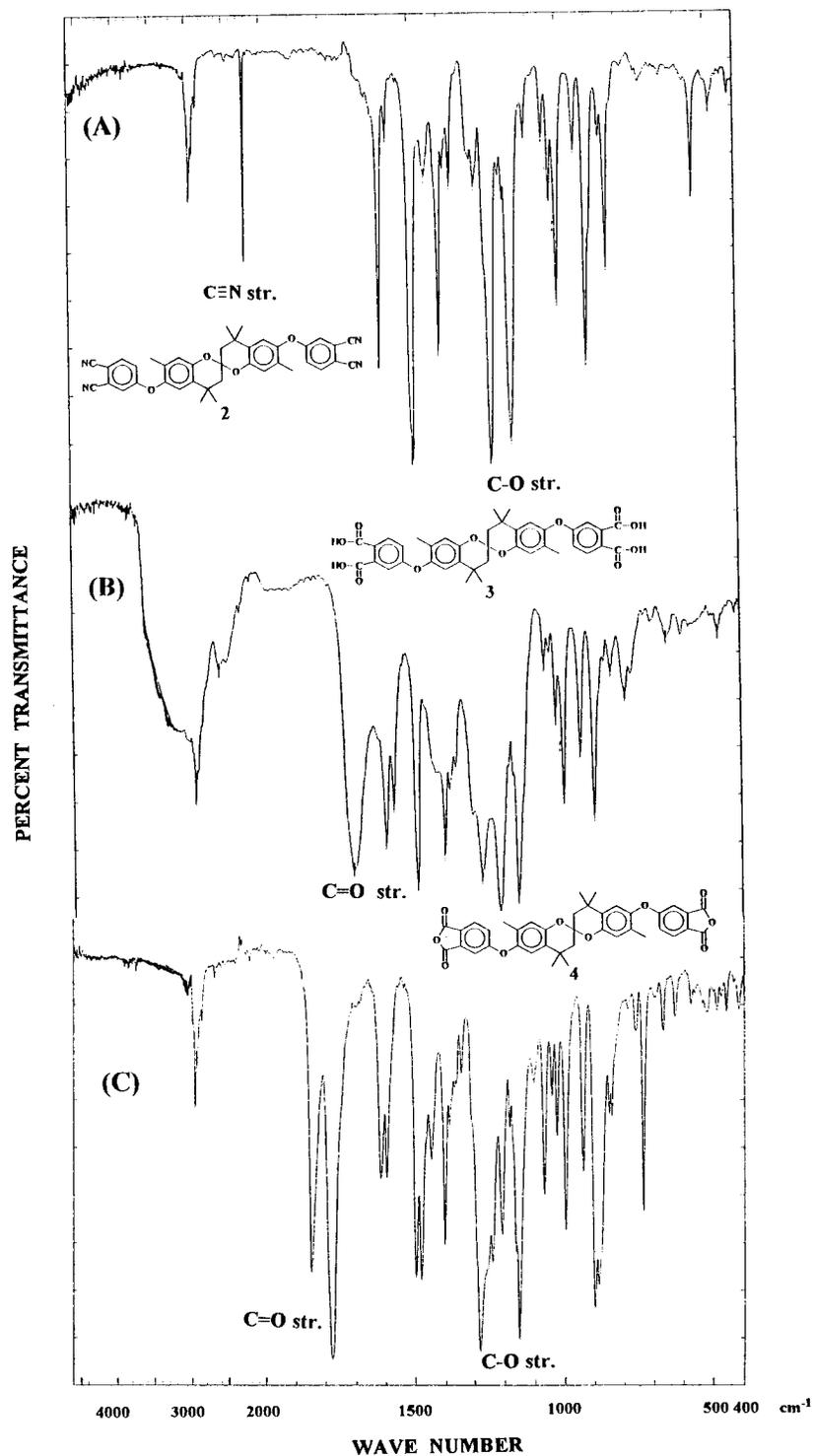


Figure 1. IR spectra of (A) tetranitrile **2**, (B) tetracarboxylic acid **3**, and (C) SBCDA (**4**).

thermal analysis station. Measurements were performed with 9–11 mg samples heated in flowing nitrogen or air of 50 cm³/min at a heating

rate of 20°C/min. The wide-angle X-ray scattering measurements were performed with a Rigaku Geiger Flex D-Max IIIa X-ray diffractometer dif-

fractometer, using Ni-filtered Cu K α radiation (40 kV, 15 mA) and the scanning rate of 2°/min. Tensile properties were determined from stress-strain curves obtained by an Instron Model 1130 universal tester with a load cell of 5 kg. A gauge of 2 cm and a crosshead speed of 5 cm/min were used. The measurements were performed at room temperature using solution-cast film specimens (0.5 cm wide, 6 cm long, and about 0.1 mm thick), and at least five individual determinations were used to form averages.

RESULTS AND DISCUSSION

Monomer Synthesis

According to the method reported in the literature,^{14,20–25} SBCDA (**4**) was prepared by a nucleophilic nitrodisplacement of 4-nitrophthalonitrile with HMSBC-diol (**1**) in which K₂CO₃ was used as the base to form the phenolate *in situ*, followed by alkaline hydrolysis of the resulting tetranitrile **2** and dehydration of the tetraacid **3** (Scheme 1). The dianhydride structure was confirmed through elemental analysis and IR spectroscopy. The IR spectra of intermediates **2** and **3** and dianhydride **4** are summarized in Figure 1. The cyano group in compound **2** absorbed near 2230 cm⁻¹. While **2** was hydrolyzed into **3**, the band for the C≡N

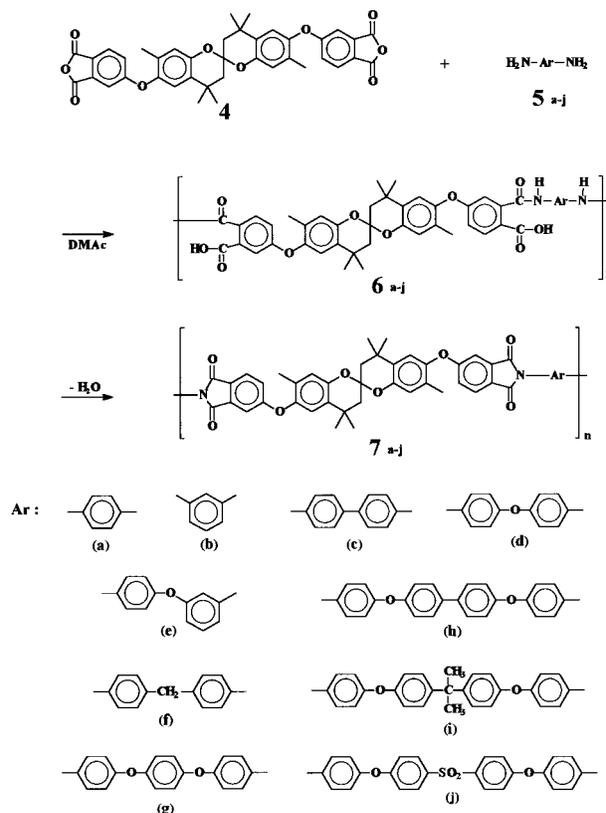
Table I. Inherent Viscosities of Poly(ether amic acid)s and Poly(ether imide)s Based on SBCDA

Poly(ether amic acid)		Poly(ether imide) ^c	
Code	η_{inh}^a (dL/g)	Code	η_{inh}^a (dL/g)
6a	0.90	7a	
6b	0.94	7b	0.46
6c	0.93	7c	0.45 ^b
6d	0.94	7d	0.59
6e	0.78	7e	0.78
6f	0.81	7f	0.60
6g	1.02	7g	0.73
6h	1.50	7h	1.28
6i	0.89	7i	0.92
6j	0.70	7j	0.60

^a Measured at a concentration of 0.5 g/dL in DMAc at 30°C using an Ubbelohde viscometer.

^b Measured in NMP using a Cannon-Fenske viscometer.

^c Obtained from thermal cyclodehydration of the poly(ether amic acid) films with a heating program: 120°C/20 min, 150°C/30 min, 200°C/30 min, and 250°C/30 min. All the poly(ether imide) films are flexible.



Scheme 2.

stretching vibration disappeared, and then appeared the broad O—H absorption in the region of 2500–3500 cm⁻¹ and the C=O stretching absorption near 1711 cm⁻¹. After compound **3** was dehydrated into dianhydride **4**, the absorption bands of O—H and C=O stretching disappeared and the characteristic absorptions of the C=O groups in cyclic anhydride were observed at 1852 and 1781 cm⁻¹.

Polymerization

Poly(amic acid)s with inherent viscosities of 0.70–1.50 dL/g (Table I) were prepared by ring-opening polyaddition of SBCDA with various aromatic diamines (Scheme 2) in DMAc at room temperature. Poly(amic acid) films were thermally cyclodehydrated to form the polyimide via stage-by-stage heating to elevated temperature. Most of the poly(ether imide) films bubbled during the imidization process, probably due to the fact that volatiles (water) were eliminated faster than they could diffuse from the films which were above T_g and bubbles formed. However, these bubbly poly-

Table II. Solubility^a of Poly(ether imide)s Based on SBCDA

Polymer	Solvent ^b								
	DMI	NMP	DMAc	DMF	DMSO	<i>m</i> -Cresol	THF	Sulfolane	Chloroform
7a	–	–	–	–	–	–	–	–	–
7b	+	+	+	+	–	+	+	–	+
7c	+	+	–	–	–	+	–	–	+
7d	+	+	+	+	–	+	+	–	+
7e	+	+	+	+	–	+	+	–	+
7f	+	+	+	+	–	+	+	–	+
7g	+	+	+	+	–	+	+	–	+
7h	+	+	+	+	–	+	+	–	+
7i	+	+	+	+	–	+	+	–	+
7j	+	+	+	+	–	+	+	–	+

^a +, soluble at room temperature; –, insoluble even on heating.

^b DMI, 1,3-Dimethyl-2-imidazolidone; NMP, *N*-methyl-2-pyrrolidone; DMAc, *N,N*-dimethylacetamide; DMF, *N,N*-dimethylformamide; DMSO, dimethyl sulfoxide; THF, tetrahydrofuran.

(ether imide) films could be redissolved in DMAc and solution-cast into smooth, flexible, and transparent films. Except for poly(ether imide) **7a**, the other poly(ether imide)s are soluble in DMAc or NMP and are characterized with inherent viscosities in the 0.45–1.28 dL/g range. The IR spectra of the polyimides have characteristic imide absorbances near 1780 and 1725 cm⁻¹ (coupled carbonyls), 1380 cm⁻¹ (C–N stretching), and 1100 and 720 cm⁻¹ (cyclic structures).

Properties of Polymers

Table II gives the results of qualitative solubility tests of these poly(ether imide)s. The poly(ether

imide) (**7a**) derived from *p*-phenylenediamine was insoluble in all the testing solvents. This may be associated with the presence of the rigid coplanar 4,4-bis(phthalimido)phenylene moiety which may lead to a higher chain packing density. However, the poly(ether imide) (**7c**) derived from another rigid diamine, benzidine (**5c**), was soluble in DMI, NMP, *m*-cresol, and chloroform. The higher solubility of **7c** when compared to **7a** may be due to the fact that although the bond between two *p*-phenylene rings in the benzidine moiety is rigid, it still has some mobility that permits a small amount of rotation of one ring with respect to the other; the rotational angle is on the order of 10–15° under the steric effects of substituents.

Table III. Tensile Properties of Poly(ether imide) Films^a

Polymer	Strength at Yield Point (MPa)	Strength at Break Point (MPa)	Elongation-to-Break (%)	Initial Modulus (GPa)
7a		102	8	2.0
7b		88	7	1.9
7c		76	5	2.0
7d	83	74	11	1.7
7e		82	8	1.8
7f		76	8	1.6
7g	78	69	15	1.8
7h	86	80	26	1.9
7i	79	71	24	1.7
7j		81	8	1.8

^a Except for **7a,c**, other testing films were cast from polyimide solutions in DMAc.

Table IV. Thermal Properties of SBCDA–Poly(Etherimide)s

Polymer	T_g^a (°C)	Decomposn. ^b Temp. (°C)		Char ^c Yield (%)
		In N ₂	In Air	
7a	214	450	456	30.3
7b	188	446	450	45.0
7c	262	457	463	40.6
7d	189	456	469	36.9
7e	175	454	459	36.8
7f	235	454	457	40.9
7g	179	460	464	36.9
7h	252	463	475	40.2
7i	200	462	468	33.7
7j	244	455	463	38.7

^a Midpoint of base line shift in the second heating DSC trace, with a heating rate of 10°C/min in nitrogen.

^b Temperature at which 10% weight loss was recorded by TG at a heating rate of 20°C/min.

^c Residual weight % at 800°C in nitrogen.

The other poly(ether imide)s were readily soluble in all the testing solvents at room temperature, except in DMSO and sulfolane. The main reason for the high solubility associated with these poly-

(ether imide)s is probably that the introduction of the bulky, nonlinear spirobichroman structure into the polymer backbone disrupts chain packing and facilitates less polymer–polymer interaction.

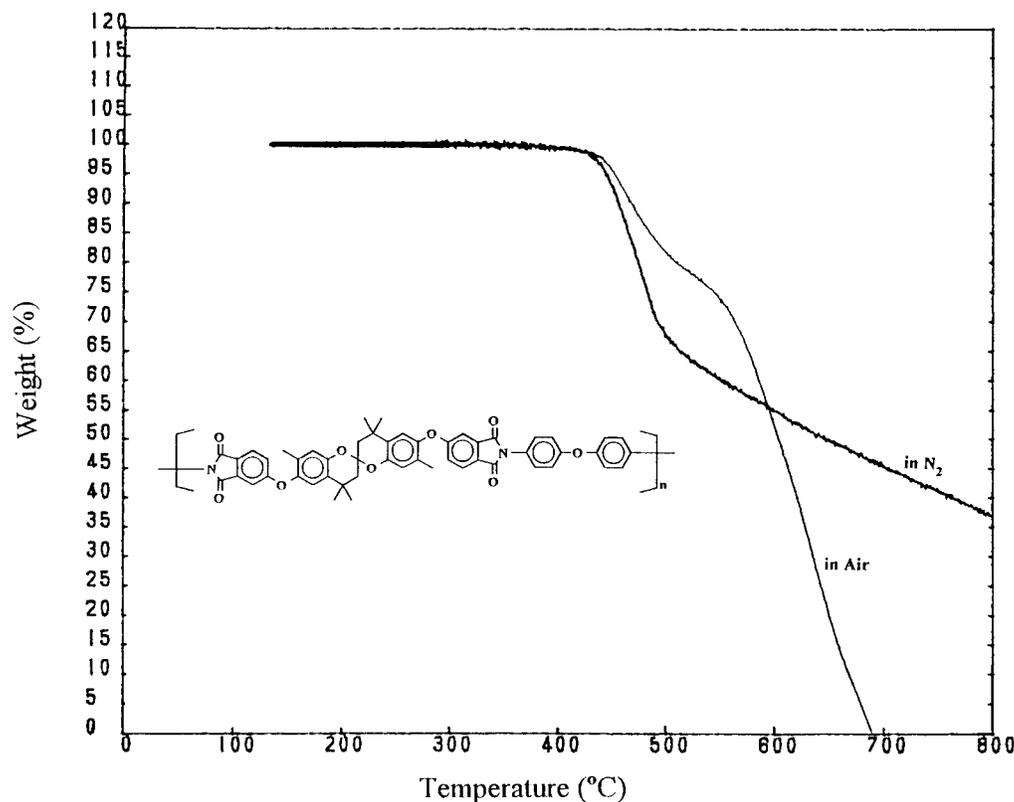


Figure 2. TG curves of polyetherimide **7d** in air and nitrogen at a heating rate of 20°C/min.

Furthermore, the high solubility of these polymers also indicates that they might be produced using the one-step method, i.e. the polymerization might be carried out at elevated temperatures in a refluxing solvent such as *m*-cresol.

The development of crystallinity for these polymers was not expected because of the spiro structure, and this was confirmed by wide-angle X-ray scans. All the poly(ether imide)s afforded flexible and noncreasable films. These films were subjected to tensile test, and their tensile properties are summarized in Table III. The films possessed a tensile strength of 69–102 MPa, elongation at break of 5–26%, and initial modulus of 1.6–2.0 GPa. Some polymer films such as those of **7d,g,h,i** behaved as ductile materials with a yield point in their stress–strain curves and moderate elongation-to-break.

The thermal properties of these poly(ether imide)s were determined by means of dynamic thermogravimetry (TG) and differential scanning calorimetry (DSC). Some thermal behavior data are listed in Table IV. As a representative example, the TG curves of the poly(ether imide) (**7d**) obtained from SBCDA and 4,4'-oxydianiline are shown in Figure 2. As seen in Figure 2, extensive degradation occurs at temperatures higher than 440°C in a nitrogen or air atmosphere. In nitrogen, the char yield at 800°C is about 37%. In air, decomposition of polymer **7d** proceeds through two steps. The first decomposition occurs at 440–500°C, and the second at 550–690°C. It appears that the polymer first loses its alicyclic segments and then undergoes degradation of the remaining aromatic segments. Moreover, it also can be seen from these TG thermograms that the rate of decomposition in air is lower than in nitrogen in the temperature range of 440–500°C. (Note: Some polymers with aliphatic segments are known to gain weight as they are oxidized and then rapidly lose weight as they degraded.) All the other poly(ether imide)s showed a similar thermal behavior, and some of their TG data are summarized in Table IV. The temperature of 10% weight loss stays in the range 446–463°C in nitrogen and 450–475°C in air. The char residue remaining at 800°C in a nitrogen atmosphere is between 30.3 and 45.0%.

DSC measurements were conducted with a heating rate of 10°C/min in flowing nitrogen. Rapid cooling from an elevated temperature (above 350°C) to room temperature yielded predominately amorphous samples so that in all

cases distinct baseline shifts were observed in the subsequent second heating traces of DSC. The midpoint of the baseline shift is defined as T_g of the polymer. As shown in Table IV, the T_g values of the poly(ether imide)s were recorded in the range 175–262°C, depending on the diamines used. In general, incorporation of a *m*-phenylene unit (e.g. in cases of **7b,e**) or an ether group (e.g. **7d,e,g**) led to a decrease in T_g , while introduction of a biphenylene unit (**7c,h**) or a sulfone group (**7j**) resulted in a higher T_g . Although not studied as part of this work, these polyimides may exhibit high permeabilities/selectivity due to the presence of spirobichroman units and therefore may prove useful in gas separation membrane applications.

CONCLUSIONS

High-molar-mass poly(ether imide)s have been obtained from SBCDA and aromatic diamines. These polymers were characterized by high solubility even in the fully imidized form, good film-forming ability, a wide temperature range between T_g and decomposition temperature, and reasonable thermal stability together with good mechanical properties. Thus, these polymers may be the candidates for high-performance engineering plastics.

We are grateful to the National Science Council of the Republic of China for the support of this work.

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ERRATUM

Synthesis and Evaluation of Novel Polyimides Derived from Spirobichroman Diether Anhydride

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On page 2807, Table IV, the second column, the T_g values of almost all polymers were incorrectly stated. This column should read as follows.

Polymer	T_g (°C)
7a	290
7b	262
7c	295
7d	257
7e	246
7f	255
7g	245
7h	260
7i	242
7j	244

On page 2801, Abstract, line 14, the paper incorrectly stated “glass transition temperatures in the range 175–262°C.” This should read “glass transition temperatures in the range 242–295°C.”

Also on page 2808, column 2, line 6, the range was incorrectly stated. The range should read “242–295°C” instead of “175–262°C.”

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