

# Synthesis and Properties of Novel Naphthalene-Containing Bismaleimides

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## SYNOPSIS

New aromatic bismaleimides containing bis(phenoxy)naphthalene unit, were synthesized. They were prepared by reacting a diamine-containing naphthalene and maleic anhydride by the usual two-step procedure that included ring-opening addition to give bismaleamic acid, followed by cyclodehydration to bismaleimide. The monomers were characterized by infrared (IR), proton nuclear magnetic resonance (<sup>1</sup>H-NMR), elemental analyses (EA), and mass spectra (MS). Their thermal polymerization was investigated by differential scanning calorimetry (DSC). The presence of a naphthalene group in the backbone of the bismaleimide had increased the curing temperature and reduced the reactivity of the maleimide bond. The glass transition temperatures of the polymers were in the range of 291–334°C. Thermogravimetric analyses of the cured resins showed higher thermal stability and char yield for naphthalene-containing bismaleimides than for the corresponding benzene analog. The observed char yield of the bismaleimide resins are in accordance with the calculated C/H ratios. © 1996 John Wiley & Sons, Inc.

## INTRODUCTION

Bismaleimides (BMI) are a class of thermosetting resins well known for their high thermal stability, hot-wet strength and fatigue resistance.<sup>1</sup> The double bond in bismaleimide is highly electron deficient due to the two flanking imide carbonyl groups. As a result, BMI resins can be thermally self-polymerized through their reactive maleic double bonds to give a highly crosslinked network without the formation of volatile by-products, offering considerable advantage in processing over the conventional condensation-type polyimide. The network exhibits chemical stability and thermal and mechanical properties superior to most epoxy resins.<sup>2,3</sup> They have become one of the important high-performance thermosetting engineering plastics in various applications, such as multilayer printed circuit boards for large-scale computers,<sup>4-6</sup> advanced composites for aerospace industries,<sup>7,8</sup> structural adhesives, and potting resin.

Although many studies have been devoted to bismaleimides and their polymers, few concern bis-

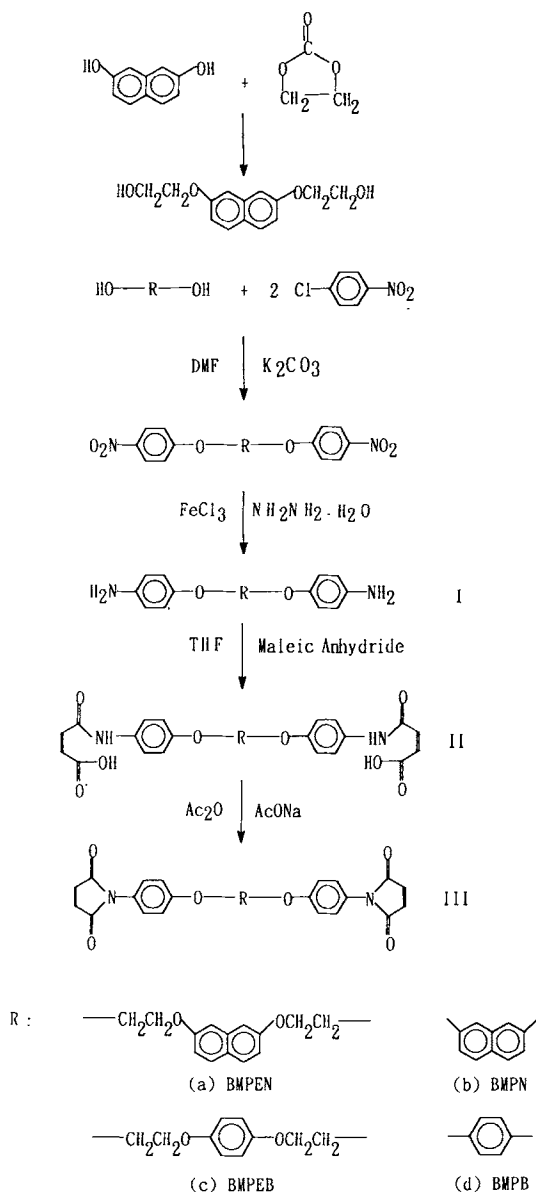
maleimides containing naphthalene structure.<sup>9-10</sup> The present work is mainly concerned with the synthesis and curing of novel bismaleimides that have naphthalene with or without an oxyethylene linkage (CH<sub>2</sub>CH<sub>2</sub>—O). The introduction of a naphthalene ring into a bismaleimide monomer structure was for the purpose of producing good thermal stability, excellent chemical resistance, and lower moisture absorption. The presence of oxyethylene bond in the backbone is expected to enhance the flexibility and the elongation at break of these resins. The thermal polymerization reactivity of the naphthalene containing bismaleimides, along with the effect of their structure on thermal stability and thermal decomposition, was investigated.

## EXPERIMENTAL

### Materials

2,7-Dihydroxynaphthalene (from Aldrich), *p*-chloronitrobenzene, hydroquinone, ethylene carbonate, potassium iodide, 1,4-bis(2-hydroxyethoxy)benzene (from TCI), ferric chloride hexahydrate (from ISHIZU), hydrazine monohydrate (from KATAYAMA),

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**Table I** Characterization of Diamines

Sample No.	Molecular Formula	Melting Point (°C)	Elemental Analysis Found (calc'd) (%)			MS ( <i>m/e</i> ) (Relative Intensity, %)
			C	H	N	
1a	C <sub>26</sub> H <sub>26</sub> N <sub>2</sub> O <sub>4</sub>	178~179	72.42 (72.56)	6.22 (6.05)	6.33 (6.51)	430 (39; M <sup>+</sup> ), 108 (52; NH <sub>2</sub> C <sub>6</sub> H <sub>6</sub> O <sup>+</sup> ) 92 (10, NH <sub>2</sub> C <sub>6</sub> H <sub>6</sub> <sup>+</sup> ).
1b	C <sub>22</sub> H <sub>18</sub> N <sub>2</sub> O <sub>2</sub>	166~167	76.97 (77.19)	5.26 (5.26)	8.18 (8.19)	342 (100; M <sup>+</sup> ), 108 (32; NH <sub>2</sub> C <sub>6</sub> H <sub>6</sub> <sup>+</sup> ).
1c	C <sub>22</sub> H <sub>24</sub> N <sub>2</sub> O <sub>4</sub>	201~202	69.39 (69.47)	6.41 (6.32)	7.32 (7.37)	380 (100; M <sup>+</sup> ), 108 (82; NH <sub>2</sub> C <sub>6</sub> H <sub>6</sub> O <sup>+</sup> ) 92 (17, NH <sub>2</sub> C <sub>6</sub> H <sub>6</sub> <sup>+</sup> ).
1d	C <sub>18</sub> H <sub>16</sub> N <sub>2</sub> O <sub>2</sub>	176~176	73.68 (73.97)	5.33 (5.26)	8.23 (8.19)	292 (100; M <sup>+</sup> ), 108 (33; NH <sub>2</sub> C <sub>6</sub> H <sub>6</sub> O <sup>+</sup> ) 92 (5, NH <sub>2</sub> C <sub>6</sub> H <sub>6</sub> <sup>+</sup> ).

maleic anhydride (from Merck), were all used as received. *N,N'*-Dimethylformamide (DMF) was purified by distillation under reduced pressure over calcium hydride and stored over 4 Å molecular sieves. All other solvents were obtained from various commercial sources and used without further purification.

## Monomer Synthesis

### 2,7-Bis(2-hydroxyethoxy)naphthalene

2,7-Bis(2-hydroxyethoxy)naphthalene was prepared from 2,7-dihydroxynaphthalene and ethylene carbonate by a modification of the method of Kem et al.,<sup>11</sup> as indicated in Scheme 1. Into a round-bottom glass flask equipped with a stirrer, a reflux condenser, a thermometer, and a nitrogen inlet were introduced 32 g (0.2 mol) of 2,7-dihydroxynaphthalene, 36.96 g (0.42 mol) of ethylene carbonate, and 0.4 g of potassium iodide. The flask was flushed with nitrogen and heated to about 100°C. Agitation was commenced as soon as the contents were melted and CO<sub>2</sub> evolution was noticed immediately. The temperature was raised to 160°C and maintained at that temperature until CO<sub>2</sub> evolution subsided (6 h). Upon cooling, the content was recrystallized from isopropanol to give a crystalline solid (38.3 g, 77.2% yield); mp 142 ~ 144°C; IR(KBr) cm<sup>-1</sup>: 3600–3200 (OH), 1600, 1510 (aromatic ring), 1450 (CH<sub>2</sub>), 1245 (C—O—C); mass spectrum EI *m/e* intensity: 248 (100, M<sup>+</sup>).

ANAL.: Calcd. for C<sub>14</sub>H<sub>16</sub>O<sub>4</sub>: C, 67.74%; H, 6.45%. Found C, 68.81%; H, 6.44%.

### General Procedure for the Synthesis of Diamine Compounds

Into a reactor equipped with a stirrer, a reflux condenser, a thermometer, a Dean-Stark water sepa-

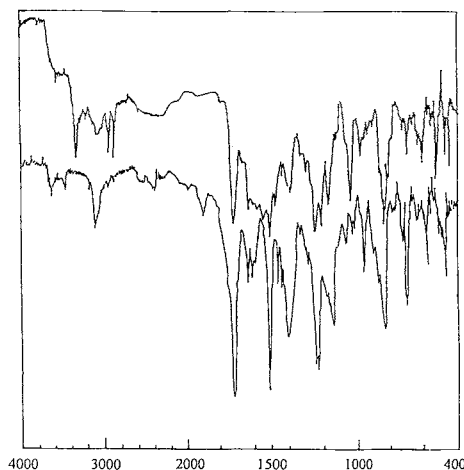
**Table II Characterization of Bismaleimides**

Bismaleimide	Molecular Formula	Elemental Analysis Found (Calc'd) (%)			IR (cm <sup>-1</sup> )				<sup>1</sup> H-NMR $\delta$ (ppm) <sup>a</sup>
		C	H	N	Imide Ring C=O	C-N-C	C-O-C	Maleimide Ring C=C	
BMPEN	C <sub>34</sub> H <sub>28</sub> N <sub>2</sub> O <sub>8</sub>	68.80 (69.15)	4.75 (4.41)	4.96 (4.75)	1775, 1710	1395	1240	680	4.4 (s, <sup>b</sup> 8H, CH <sub>2</sub> ); 7.0-7.8 (m, <sup>c</sup> 18H, CH=CH, and ar <sup>d</sup> )
BMPN	C <sub>30</sub> H <sub>18</sub> N <sub>2</sub> O <sub>6</sub>	70.94 (71.71)	3.88 (3.59)	5.51 (5.58)	1780, 1720	1390	1250	690	7.15 (s, 4H, olefinic); 7.18-8.00 (m, 14H, ar)
BMPEB	C <sub>30</sub> H <sub>24</sub> N <sub>2</sub> O <sub>8</sub>	66.59 (66.67)	4.54 (4.44)	5.26 (5.19)	1785, 1710	1395	1250	680	4.2-4.3 (s, 8H, CH <sub>2</sub> ); 6.9 (s, 4H, olefinic); 7.0-7.3 (m, 12H, ar)
BMPB	C <sub>26</sub> H <sub>16</sub> N <sub>2</sub> O <sub>6</sub>	68.87 (69.03)	3.62 (3.54)	6.22 (6.19)	1775, 1720	1400	1245	690	—

<sup>a</sup> In DMSO-d<sub>6</sub>.

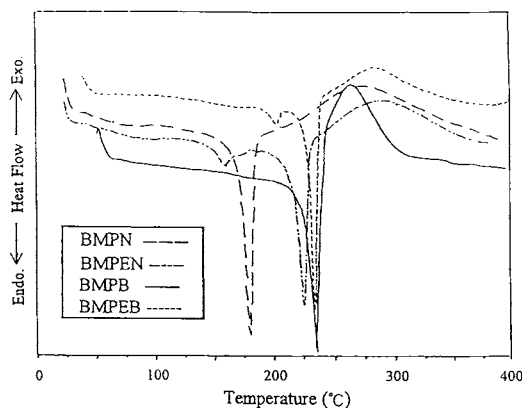
<sup>b</sup> Singlet.

<sup>c</sup> Multiplet.

<sup>d</sup> Aromatic.

**Figure 1** IR spectra of bismaleamic acid IIa (top) and the bismaleimide BMPEN (bottom).

rator, and a nitrogen inlet were charged 250 mL of DMF, 0.3 mol of dihydroxy monomer, 0.66 mol of *p*-chloronitrobenzene, 82.8 g (0.6 mol) of potassium carbonate, and 75 mL of toluene. The reaction mixture was refluxed at 140–145°C for 8 ~ 12 h under a nitrogen atmosphere. Water produced by the reaction was successively removed from the system by azeotropeing with toluene. After completion of the reaction, the resulting inorganic salts were removed by hot filtration. These salts were washed with 90 mL of DMF, then the combined filtrate was heated again to 100–110°C. To the mixture, 60 mL of water was added and slowly cooled to room temperature. The crude product precipitated gradually; the precipitate was collected by filtration. The solids were washed with 500 mL of methanol, filtered, and dried under vacuum.

To 0.1 mol of the dinitro compound in 200 g of ethanol was added 2 g of charcoal and 0.3 g of ferric chloride-hexahydrate. Over a period of 1 h, 50 mL of 85% hydrazine monohydrate was added to the


**Figure 2** DSC scans of bismaleimides.

above mixture at 85°C and then maintained at that temperature for 4 h. The product (yield 95%) was recrystallized from 2-methoxyethanol or DMF.

### General Procedure for the Synthesis of Bismaleimide Compounds

A three-necked flask equipped with an addition funnel and a nitrogen inlet was charged with a solution of appropriate diamine (0.1 mol) in THF (100 mL). Under nitrogen atmosphere, maleic anhydride (21.56 g, 0.22 mol) dissolved in THF (50 mL) was added dropwise to the above mixture. An exothermic reaction occurred and product precipitation was observed immediately. The mixture was stirred for 4 h at room temperature and then at 60°C for an additional 1 h. The yellow precipitate was collected, washed with fresh THF to remove excess maleic anhydride, and then dried in vacuum.

To a 500-mL round-bottomed flask was added 26.9 g (0.05 mol) of bismaleamic acid and 100 mL of DMF. The solution was heated to 60–65°C with stirring. Acetic anhydride (30 mL) and 2.7 g of sodium acetate (300 mL and 27 g, respectively, per mole of water condensed)<sup>12</sup> were added to the preheated solution. The temperature of reaction mixture was raised to 80–90°C and then stirred at that temperature for 6 h. The solution was cooled to room temperature with constant stirring and then poured into ice water with stirring for 1 h. The precipitate was filtered and washed with aqueous sodium carbonate. A yellow solid (yield 96%) was obtained.

### Measurements

Melting points were obtained on a polarizing microscope (Laboratory Devices MEL-TEMP. II.) melting-point apparatus and were uncorrected. Infrared (IR) spectra were recorded on a Jasco IR-810 spectrometer with KBr pellets. Nuclear magnetic resonance (NMR) spectra were obtained using a Bruker WP-100 spectrometer at 200 MHz with DMSO-*d*<sub>6</sub> as solvent and tetramethylsilane as an

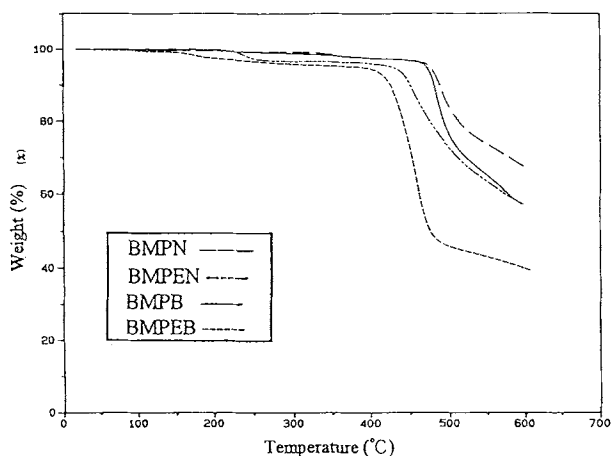


Figure 3 TGA thermograms of cured bismaleimides.

internal standard. Elemental analyses were carried out with Heraeus CHN-Rapid elemental analyzer. Mass spectrometric analyses were performed on a VG 70-250 S GC/MS spectrometer with a solid inlet. DSC data were obtained from 8–10-mg samples in a nitrogen atmosphere at a 20°C/min heating rate using a Du Pont 910 differential scanning calorimeter. Thermogravimetric analyses (TGA) were performed on a Du Pont 951 thermal analyzer, using a heating rate of 20°C/min in *N*<sub>2</sub> at a flow rate of 40 cm<sup>3</sup>/min. Dynamic mechanical analyses (DMA) were made with a Perkin-Elmer DMA7e. The storage modulus *G'* and tan  $\delta$  were studied when the sample was subjected to temperature scan mode with a three-point bending system at a programmed heating rate of 5°C/min from ambient to 350°C at a frequency of 1 Hz.

## RESULTS AND DISCUSSION

### Monomer Synthesis

The synthetic procedure described by Kem<sup>11</sup> was used to prepare the 2,7-bis(2-hydroxyethoxy)-naphthalene. Liquid chromatography was used to

Table III Results of Thermal Analysis of Various Bismaleimides

	$T_m^a$ (°C)	$T_i^b$ (°C)	$T_{exo}^c$ (°C)	$T_i - T_m$ (°C)	$T_g^d$ (°C)
BMPEN	226	233	289	7	316
BMPN	181	227	274	46	334
BMPEB	234	244	286	10	291
BMPB	236	—	265	0	323

<sup>a</sup> Melting point from DSC with a heating rate of 20°C/min.

<sup>b</sup> Onset temperature for curing reaction.

<sup>c</sup> Maximum temperature of polymerization exotherm from DSC with a heating rate of 20°C/min.

<sup>d</sup> Glass transition temperature from DMA.

**Table IV Results of Thermogravimetric Analysis of Uncured Bismaleimides**

	$T_{10\%}^a$ (°C)	$T_{max}^b$ (°C)	Char Yield at 600°C (%)	C/H Ratio (Calc'd.)
BMPN	479	484	66	1.67
BMPEN	435	442	59	1.31
BMPB	465	478	63	1.63
BMPEB	411	431	41	1.25

<sup>a</sup> Temperature at which 10% weight loss was recorded by means of thermogravimetry at a heating rate of 20°C/min.

<sup>b</sup> Maximum polymer decomposition temperature.

follow the reaction, however, the subsidation of carbon dioxide evolution was a convenient indicator for the completion of the reaction. Faster reaction rates were observed at higher temperature. However, decomposition of product and production of by-product were likely to occur at the higher temperature. The optimum temperature was found to be around 160°C.

The diamine containing a naphthalene ring and oxyethylene linkage was obtained from 2,7-dihydroxynaphthalene according to synthetic Scheme 1. The dinitro compound was synthesized in a quantitative yield from 2,7-dihydroxynaphthalene and *p*-chloronitrobenzene in the presence of potassium carbonate dissolved in dry DMF and refluxed for 8 ~ 12 h. Reduction of the dinitro compounds to amino compounds was carried out with hydrazine/ $FeCl_3$ . The disappearance of characteristic nitro stretching bands at around  $1510 \pm 10$  and  $1335 \pm 10$   $cm^{-1}$  on the IR spectra was used as criteria for the completion of the reduction of dinitro compounds. The structures of the intermediate dinitro and diamine compounds were confirmed by IR,  $^1H$ -NMR, mass spectra, and elemental analyses. Electron impact induced fragmentation patterns of these diamines at 30 eV have been obtained. Molecular ion peak from Ia to Id were observed at 430, 342, 380, and 292 with relative intensities of 29, 100, 100, and 100 for base peaks, respectively. The scission of the ( $NH_2-Ph-O-$ ) group led to the intense  $m/z$  108 fragment ion peak, which was observed in all mass spectra of these diamines. The results of elemental analyses of these diamine compounds in Table I were all in agreement with the calculated values for the proposed structures.

The diamines were reacted with the respective amount of maleic anhydride in THF at ambient to give the bismaleamic acids. The reaction was exothermic and the product precipitated immediately upon mixing of the reagents. The presence of bismaleamic acids was noticed from  $^1H$ -NMR spectra

**Table V Results of Thermogravimetric Analysis of Cured Bismaleimides**

	$T_{10\%}^a$ (°C)	$T_{max}^b$ (°C)	Char Yield at 600°C (%)
BMPN	487	491	67
BMPEN	451	458	57
BMPB	481	484	57
BMPEB	425	457	40

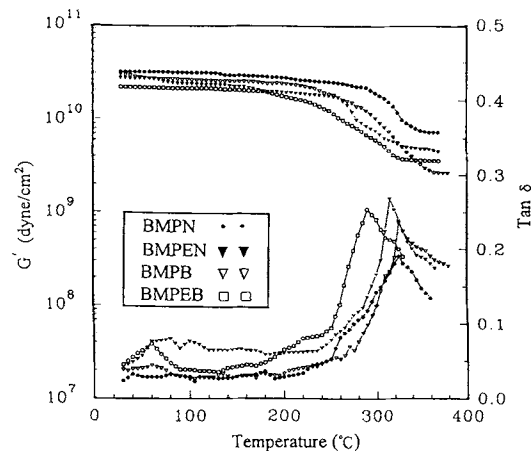
Cured at 250°C for 4 h and postcured 280°C for 0.5 h.

<sup>a</sup> Temperature at which 10% weight loss was recorded by means of thermogravimetry at a heating rate of 20°C/min.

<sup>b</sup> Maximum polymer decomposition temperature.

that showed the absence of amino protons of the diamine and the presence of four protons at 6.25–6.50 ppm corresponding to the *cis* olefinic protons. Also observed was a singlet for two carboxylic protons at 10.4 ppm. The integration matched the expected spectra. This confirmed the attachment of the maleic anhydride unit at the amino terminal to give bismaleamic acid (II). The infrared spectra of bismaleamic acids (II) showed the presence of carboxylic group ( $1720 \pm 10$   $cm^{-1}$ ) and amide group ( $3300$  and  $1540$   $cm^{-1}$ ).

The bismaleamic acids were cyclodehydrated by moderate heating utilizing acetic anhydride and sodium acetate in DMF. The heterogenous mixture became a homogeneous solution at the end of imidization for BMPN and BMPEN. The typical infrared spectra of BMPEN bismaleimide (IIIa) and bismaleamic acid (IIa) exhibited several distinctive differences as shown in Figure 1. All bismaleimides showed a strong absorption at  $1720 \pm 10$   $cm^{-1}$  and weak absorption at  $1780 \pm 10$   $cm^{-1}$ , assigned respectively to asymmetrical and symmetrical  $C=O$  stretching vibration of imide ring, and lacked the

**Figure 4** DMA results for the bismaleimide resins.

absorption at  $3400\text{--}3000\text{ cm}^{-1}$  associated with the carboxylic OH stretching vibration and  $1540 \pm 10\text{ cm}^{-1}$  for the  $\text{—NH}$  stretching of the amide group. Consequently imidization could be monitored by means of these absorptions. However, interpretable  $^1\text{H-NMR}$  spectra of BMPB bismaleimide could not be recorded because of its insolubility, even in polar aprotic solvents. The other bismaleimides were characterized by  $^1\text{H-NMR}$  spectroscopy; a sharp singlet at  $\delta$  7.15 or 6.9 ppm due to the olefinic protons of the bismaleimides was observed, while the carboxylic protons of bismaleamic acids had disappeared. The bismaleimides synthesized are characterized in Table II.

### Polymerization of Monomers

DSC traces of the four bismaleimides are given in Figure 2, and the values of transition temperature and curing characteristics are reported in Table III. A sharp endothermic peak is due to the melting of these bismaleimides. All bismaleimides exhibited an exothermic transition associated with curing in the temperature range  $220\text{--}290^\circ\text{C}$ . BMPB is polymerized immediately after being melted, because of the small difference between the melting point and the onset temperature for the curing reaction  $T_i$ . As a result, it is difficult to maintain the uncured resins in a fluid state. During the polymerization process, the viscosity of the resin quickly increased which trapped air bubbles easily and led to process difficulties, while naphthalene-containing monomer (BMPN) had a larger processing window than BMPB with good fluidity and processability. The curing reactivity of bismaleimides may be judged by  $T_{\text{exo}}$  values. The  $T_{\text{exo}}$  value decreased in the following order  $\text{BMPEN} > \text{BMPEB} > \text{BMPN} > \text{BMPB}$ . BMPEN and BMPN with naphthalene rings in the backbone required higher curing temperature than the corresponding benzene analog because of their reduced reactivity. The steric hindrance of the naphthalene ring is larger than that of benzene ring. During the thermal polymerization, the mobility of reactive sites is restricted. The lower diffusion rate resulted in slower polymerization and the formation of crosslinked cured product.<sup>13</sup> Bismaleimides BMPEN and BMPEB with extended oxyethylene chains required higher cure temperatures relative to that of bismaleimides BMPN and BMPB. The exotherm was shifted to a higher temperature as the length and the formula weight of the bridge between the maleimide group was increased.<sup>14</sup> The  $T_g$  of the crosslinked resins could not be detected by DSC, this could be attributed to the fact that the resin

was highly crosslinked and the change in  $C_p$  was very small.

### Thermal Stability of Polymer

Thermogravimetric analysis (TGA) is the most favored technique for the rapid evaluation of the thermal stability of polymeric materials. It is especially useful in comparing and ranking the thermal stabilities of various polymers. Thermogravimetric analyses of the uncured and cured samples were carried out in a nitrogen atmosphere, and the thermogravimetric traces are given in Figure 3. The thermal stability of these resins were compared by the temperature of 10% weight loss ( $T_{10\%}$ ), temperature of maximum rate of weight loss ( $T_{\text{max}}$ ), and percentage char yield at  $600^\circ\text{C}$ . They are presented in Tables IV and V. On the basis of 10% weight loss temperature, the following order may be given for the thermal stability of bismaleimides:  $\text{BMPN} > \text{BMPB} > \text{BMPEN} > \text{BMPEB}$ . These results indicate that the naphthalene rings containing BMPN and BMPEN have higher thermal stability than the corresponding benzene analog BMPB and BMPEB. The reason may be that the resonance effect and bond dissociation energy of naphthalene ring are greater than those of the benzene ring. On the other hand, the  $T_{10\%}$  values of the resins containing oxyethylene linkage BMPEN and BMPEB were less than the  $T_{10\%}$  of the resins containing aromatic ether bonds. These results suggest that the ethylene-oxygen ( $\text{CH}_2\text{CH}_2\text{—O}$ ) bond is more liable to break in comparison with the phenyl-oxygen ( $\text{Ph—O}$ ) linkage. The slight weight loss under  $250^\circ\text{C}$  was observed in BMPEN and BMPEB, which may be due to residual moisture or low molecular weight impurities in the resins. The observed char yield are in accordance with the calculated C/H ratio. The uncured BMPN monomer with the highest C/H ratio, gave the highest char yield. The anaerobic char yield for cured resins was at a maximum in BMPN (67%). The bismaleimides BMPN and BMPEN with naphthalene rings had higher char yield than the bismaleimides containing benzene ring.

### Dynamic Mechanical Properties of Polymers

The peak of  $\tan \delta$  was identified as the glass transition temperature ( $T_g$ ) because a large decrease in  $G'$  occurred at this point. The glass transition temperatures of polymers III ranged between  $291^\circ\text{C}$  and  $334^\circ\text{C}$ . The dynamic mechanical properties of the bismaleimides resins are indicated in Figure 4. BMPEN and BMPEB which contain ethylene-oxygen ( $\text{CH}_2\text{CH}_2\text{—O}$ ) group show lower storage mod-

ulus and lower  $T_g$  than those of BMPN and BMPB which contain phenyl-oxygen (Ph-O) group. This may be attributed to the reduced crosslinking density, or to the chain flexibility of the polymer matrix.

## CONCLUSION

Novel bismaleimides containing naphthalene structure were synthesized. The introduction of the naphthalene ring into bismaleimides increased their thermal stability, while reducing their polymerization reactivity in comparison to their benzene analog. Resins with oxyethylene linkage had lower storage moduli and lower thermal decomposition temperatures than resins without it.

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