Thermal Behavior and Properties of Naphthalene Containing Bismaleimide–Triazine Resins

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ABSTRACT: A series of bismaleimide · triazine (BT) resins were prepared from various dicyanate esters and 2,7-bis(4-maleimidophenoxy)naphthalene (BMPN), which contains a naphthalene group and an aryl ether linkage in the backbone. Their curing behaviors were characterized by differential scanning calorimetry. The exotherm temperature and polymerization reactivity were strongly affected by the chemical structure of the various dicyanate ester monomer. Thermal behaviors were investigated by thermogravimetric analyses and dynamic mechanical analyses. The glass transition temperatures (T_g) of these cured resins with bismaleimide/dicyanate ester at the 1/2 molar ratio were in the range of 250–322°C, and exhibited excellent thermal stability up to 400°C in nitrogen. These results provided a structure–properties relationship for the cured bismaleimide · triazine resins. © 1998 John Wiley & Sons, Inc. J Appl Polym Sci 68: 1199–1207, 1998

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INTRODUCTION

Conventional electronic circuit boards are made from glass–fiber reinforced epoxy resins. However, they often fall short of the thermal and electrical performance demands of many modern high-speed devices. With current trends toward increased circuit densities, shorter propagation delays, elevated operating temperatures, and higher reliability, advanced materials such as bismaleimide (BMI), cyanate ester (CE), and bismaleimide · triazine (BT) resins are required to satisfy these demands.[1–3] Among these materials, bismaleimide · triazine is considered to be a very promising system. In the early 1960s, Grigat and Putter were the first to report the synthesis of stable aromatic cyanates.[4] Since then, mono-, di-, and polyfunctional cyanates as well as other derivatives have been synthesized. The first cyanate resin (Triazine A) was introduced for the circuit board application in the mid-1970s,[5] and later, Mitsubishi Gas Chemical company developed a bismaleimide · triazine (BT) resin system.[6] However, BT resins were not made available until 1985 by Hi-Tek Polymers, Inc., who marketed a series of resin systems for both circuit board and advanced composite applications.

Cyanate resins have many excellent properties, such as good thermal stabilities, high glass transition temperature, low dielectric constant over a wide range of frequency and temperature, and outstanding adhesion to conducting metals. They

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b.

c.
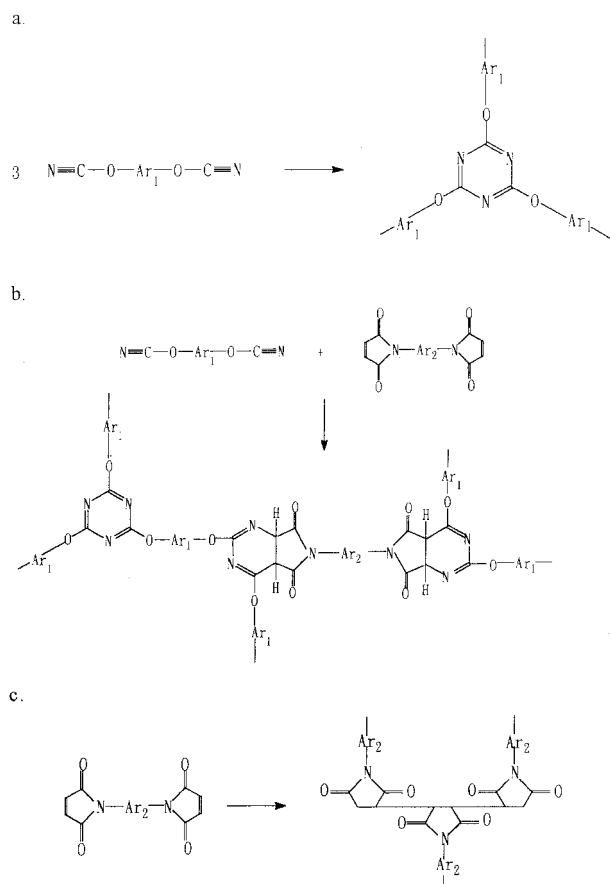
Scheme 1. Bismaleimide/triazine (BT resin) chemistry.

can be mixed with many kinds of thermosetting resins to form compatible formulation blends for aerospace and electronic applications. Aerospace applications include high-temperature adhesives, protective coatings, and matrix resins for structural components. Electronic applications include printed-wiring circuit boards, thin cards, multichip module laminates (MCM-L), and chip encapsulants.[7-9]

Mitsubishi Gas Oil Chemicals developed a new family of resins that are mixtures of difunctional cyanate ester and bismaleimide (BT resins; B stand for bismaleimide, T triazine). The major reactions upon curing of the mixture include the cyclotrimerization of three aryl cyanates (Ar—O—C≡N) to form triazine, reaction of dicyanate with bismaleimide forming pyrimidine structure, and polymerization of bismaleimide (Scheme 1 a, b, and c[10]). These resins have substantially better resistance to catastrophic loss of insulation resistance than the prevailing epoxy or bismaleimide

systems.[11,12] With superior properties, such as reduction in moisture absorption, excellent chemical resistance, good dimensional stability, low dielectric constant, and low dielectric loss, BT resins are attractive for circuit boards and semiconductor encapsulants applications for which high reliability is critical.

A wide variety of bismaleimides (BMIs) have been studied and introduced into the marketplace. Nevertheless, few concern bismaleimide containing a naphthalene structure. We have synthesized 2,7-dihydroxynaphthalene-based bismaleimide.[13,14] In our previous study,[15] crosslinking of this novel bismaleimide (2,7-bis(4-maleimidophenoxy)naphthalene, BMPN) and the commercial 4,4-bismaleimidophenyl methane (BMPM) using 4,4-diamino diphenylmethane (DDM) at the 1/2 molar ratio of bismaleimide/DDM was investigated. BMPN network that contains naphthalene exhibited higher glass transition temperature, thermal stability, modulus, and moisture resistance than that of BMPM or the 2,7-dihydroxynaphthalene-based epoxy system. The object of this research was to investigate the curing characteristics of a series of novel BT resins that were prepared from various dicyanate esters and 2,7-bis(4-maleimidophenoxy)naphthalene (BMPN). The resulted BT resins were expected to have enhanced thermal stability, excellent chemical resistance, and lower moisture absorption than commercial BT resins because of the naphthalene unit in the backbone structure. In addition, the incorporation of aryl ether linkage into the backbone will impart processability and solubility without much loss of its thermal stability. The curing characteristic was studied by DSC. The thermal properties, dynamic mechanical analysis, and moisture resistance of the crosslinked bismaleimide · triazine resins were also investigated to understand the structure–properties relationship.

EXPERIMENTAL

Material

2,7-Dihydroxynaphthalene (from Aldrich, Milwaukee, WI), maleic anhydride (from Merck), cyanogen bromide (from Aldrich), and copper acetylacetonate catalyst (from TCI) were all used as received. Various bisphenols such as hydroquinone, 4,4′-biphenol, bis(4-hydroxyphenyl)methane, 4,4′-dihydroxydi-

phenyl ether, 2,2-bis(4-hydroxyphenyl)propane, and 4,4′-methylenebis (2,6-dimethylphenol) were obtained commercially and used without further purification. Triethylamine was distilled from barium oxide; tetrahydrofuran (THF) was distilled from a mixture of Na metal and benzophenone; acetone was distilled from Drierite prior to use.

### Preparation of Bismaleimide Monomer (BMPN)

2,7-bis(4-maleimidophenoxy)naphthalene (BMPN) was prepared from maleic anhydride and 2,7-bis(4-aminophenoxy)naphthalene (BAPN) by the method described in our previous reports.[13,14] 2,7-Bis(4-aminophenoxy)naphthalene (BAPN) was synthesized according to the method reported,[13,14] m.p. 166–167°C (Scheme 2). In general, a solution of appropriate diamine (0.1 mol) in tetrahydrofuran (THF, 100 mL) was prepared. Under nitrogen atmosphere, maleic anhydride (21.56 g, 0.22 mol) dissolved in 50 mL of THF was added dropwise to the above mixture. An exothermic reaction occurred and precipitation of products 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 a vacuum.

To a 500-mL round-bottomed flask were placed 0.05 mol of bismaleamic acid and 100 mL of DMF, and the solution was heated to 60–65°C with stirring. Acetic anhydride (30 mL) and 2.7 g of sodium acetate were added to the preheated solution. The temperature of reaction mixture was raised to 80–90°C and then allowed to stir 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 was obtained. IR (KBr) cm^{-1}: 1780, 1720 (C=O, imide ring), 1390 (C—N—C), 690 (C=C, maleimide ring), ^{1}H-NMR (DMSO-d$_6$) δ (ppm): 7.15 (s, 4H, olefinic), 7.18–8.00 (m, 14H, aromatic), FAB Mass spectrum intensity (%): 502 (20, M$^+$).

ANAL Calcd. for C$_{30}$H$_{18}$N$_2$O$_6$: C, 71.71%; H, 3.59%; N, 5.58% found C, 71.67%; H, 3.75%; N, 5.42%.

General Procedure for the Preparation of Dicyanate Ester Monomers

The dicyanate monomers were synthesized using a modified procedure reported by Grigat et al.[4]
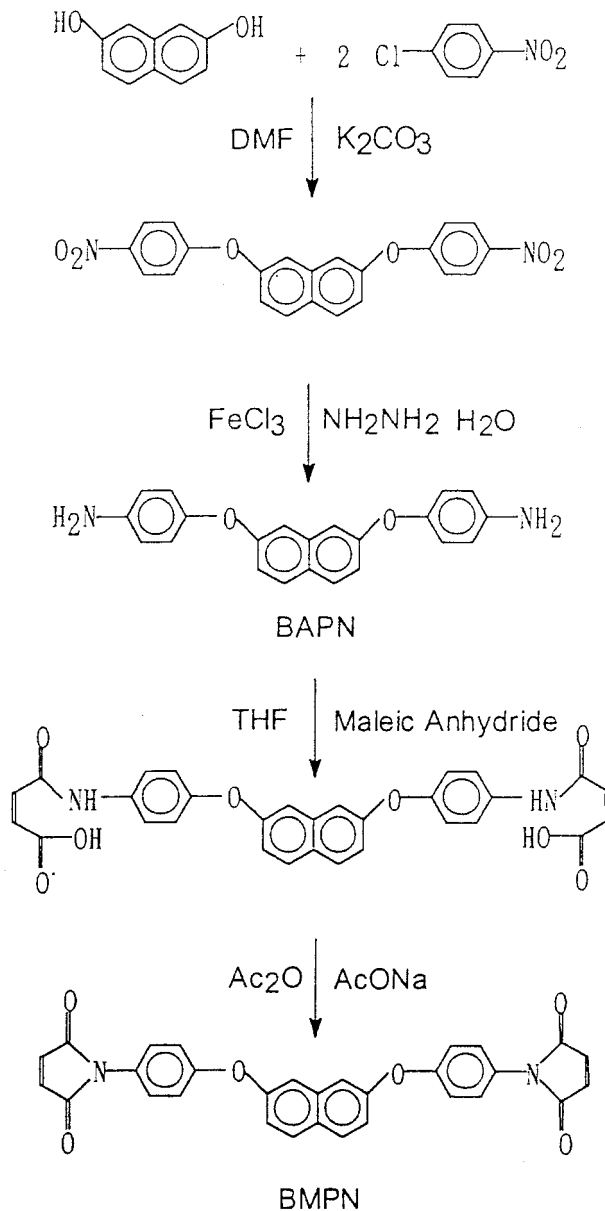
Scheme 2. Bismaleimide monomer synthesis.

(Scheme 3). To a 250-mL four-necked flask were charged bisphenols (0.05 mol), triethylamine (0.11 mol), and 30 mL of acetone or tetrahydrofuran (THF). The solution was stirred with a mechanical stirrer and maintained at −5°C. Under nitrogen atmosphere, cyanogen bromide (BrCN, 0.11 mol) in acetone (20 mL) was added dropwise into the flask over a period of 1 h at −5°C–0°C. After the addition was completed, the temperature was allowed to equilibrate to room temperature. The solution was added dropwise to ice water. The precipitate was collected and washed
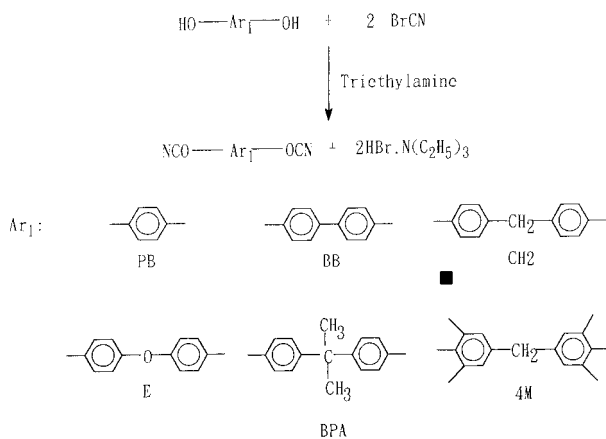
Scheme 3. Dicyanate ester synthesis.

with water. The dicyanate monomers were white crystalline powders.

Measurements

Infrared (IR) spectra were recorded on a Jusco 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_6 as solvent and tetramethylsilane as an 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_2 at a flow rate of 40 cm^3/min. Dielectric analyses were obtained using a Du Pont 9100 dielectric analyzer with two parallel plate systems, in N_2 at a flow rate of 500 cm^3/min and a frequency of $1M$ Hz. Dynamic mechanical analyses (DMA) were made with a Perkins–Elmer DMA 7e. The storage modulus G' and tan δ were studied when the sample was subjected to temperature scan mode with a three-point bending system at a heating rate of 5°C/min from ambient to 350°C at a frequency of 1 Hz.

Moisture Absorption

Disk samples [1 mm (T) × 50 mm (D)] of cured resins were exposed to boiling distilled water for 72 h. The quantity of water absorbed as a result of these exposures was determined by weight differences before and after exposures. Before each weight measurement, the samples were wiped to remove surface water and weighed immediately. The results of these tests were plotted graphically for each resin system.

RESULTS AND DISCUSSION

The synthesis of the aryl ether groups containing naphthalene has been reported in previous articles.[13,14] The cyanate ester monomer formation is described in Scheme 3 with various bisphenols. The cyanate ester compounds were synthesized from bisphenol and cyanogen bromide in the presence of triethylamine as an acid acceptor using acetone as a solvent. THF was used instead of acetone when bisphenol had poor solubility in acetone. The reaction was exothermic and cyanogen bromide was fed in slowly to maintain the reaction at the desired rate. High reaction temperature would lead to unwanted formation of diethyl cyanamide. The typical infrared spectrum of the dicyanate ester (BPA) exhibited several distinctive absorptions. The dicyanate showed strong absorptions at 2280 and 2230 cm^{-1}, assigned to cyanate C≡N stretching vibration, and lacked the absorption at 3500–3200 cm^{-1} associated with the phenolic OH stretching vibration. Consequently, the reaction could be monitored by means of these absorptions. Molecular ion of dicyanate ester (BPA) was observed at 278 with relative intensity of 22 for base peak. The scission of the methyl group led to the intense m/z 263 (M-15) fragment ion with relative intensity 100 for the base peak.

The conversion of the bisphenol monomers to their dicyanate esters was also confirmed by the disappearance of the phenolic proton in ^{1}H-NMR, and the downfield shift of the protons ortho to the —OCN functional groups. The typical infrared spectrum of BPA dicyanate ester that had been cured at 200°C for 30 min (prepolymer formation) exhibited several distinctive differences from its monomer, as shown in Figure 1. In addition, the absorptions assigned to triazine at 1580 and 1380 cm^{-1} were observed. These vibrations can be attributed to the triazine ring formation from the cyclotrimerization reaction involving the cyanate groups. The peak height of the cyanate group at 2280 and 2230 cm^{-1} decreased. The C—H stretch-
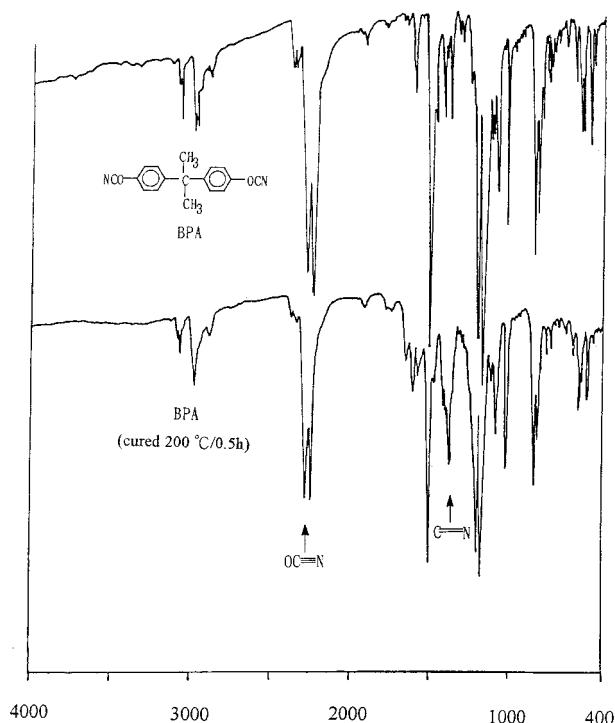
Figure 1 IR spectra of BPA cyanate ester monomer (top) and its prepolymer cured at 200°C for 30 min (bottom).

ing at 2950 cm^{-1} was used as the reference peak. The structures of the dicyanate ester monomers were confirmed by elemental analyses, mass spectra, and ^{1}H-NMR, which are presented in Table I. The results are in good agreement with the proposed structures.

Thermal Behavior

The curing behaviors of various dicyanate esters were examined by DSC. Representative thermograms of some monomers are shown in Figure 2. The endothermic transition temperature (T_m), onset temperature for curing reaction (T_i) and exothermic peak temperature (T_{exo}) are reported in Table II. All dicyanates exhibited an endothermic transition associated with melting in the temperature ranged from 83 to 141°C. All the dicyanate monomers showed a "processing window" (temperature interval between melting and curing) in the temperature ranged from 64 to 133°C, respectively, at a heating rate of 20°C/min. Thus, this wide processing window and low melting temperature enable these polymer precursors to flow at a low temperature and to crosslink at a higher

temperature, which is advantageous to the processor.

The curing reactivity of dicyanate ester may be judged by T_{exo} values. PB had the lowest T_i and T_{exo} among all dicyanate esters, and this may be attributed to its reactive end groups being the closest. Methyl substituents ortho to the reactive —OCN group in 4M monomer had shielded and sterically hindered the reactive groups and led to a restriction in the segmental mobility and thus delayed the exothermic temperature.[15]

When 500 ppm Cu acetylacetonate catalyst was added to the curing system with dicyanate ester and bismaleimide (BMPN) at a 1/2 molar ratio, the crosslinking reaction was initiated at a much lower T_i and T_{exo} temperature than without the catalyst (comparing Table II with Table III). The curing characteristics with the catalyst are given in Table III. The polymer BT$_{PB}$ had the lowest T_{exo} and, hence, the highest curing reactivity. The BT$_{4M}$ with four methyl substituents requires the highest curing temperature. From Table II and Table III, the resins containing phenyl–ether (ph—O) linkage (E, BT$_E$) had lower T_{exo} than that of resins with ph—C(CH$_3$)$_2$—, ph—CH$_2$ linkage. It may be attributed to the flexibility of phenyl–ether (ph—O) linkage, which reduced chain stiffness and introduced a rather high mobility into the chain.[16] Consequently, a lower curing temperature was observed for E and BT$_E$ with ether linkage.

Thermal Stability of Polymers

The relative thermal stability of cured resins were compared by the temperature of 5 and 10% weight loss ($T_{5\%}$ and $T_{10\%}$), temperature of maximum rate of weight loss (T_{max}), and percent char yield at 600°C (Y_c). The TGA data for cured resins at various molar ratio of BMI/dicyanate ester are listed in Table IV. These polymers exhibited 5% weight loss decomposition temperature ($T_{5\%}$) ranging between 401–475°C in nitrogen. Their maximum decomposition temperatures (T_{max}) ranged between 428 and 491°C, and the anaerobic char yields at 600°C ranged between 42 and 71%. The thermal stability of cured BT resins increased with the increase in the starting BMI/dicyanate molar ratio, which may be due to the higher crosslink density of BMI than cyanate ester.[17]

The thermal stabilities of cured BT resins with dicyanate ester and BMI molar ratio of 2/1 were presented in Table V. Representative thermo-

Table I   Characterization of Cyanate Ester Monomers

| Cyanate Esters | Elemental Analysis Found (calcd) (%) | | | ^{1}H-NMR δ^a (ppm) | MS (m/e) |
	C	H	N		
PB	60.21 (60.00)	2.42 (2.50)	17.57 (17.50)	7.64 (s^b, 4H, ar^d)	160 (M+, 100)
BB	71.34 (71.19)	3.48 (3.39)	11.71 (11.86)	7.52–7.89 (m^c, 8H, ar)	236 (M+, 100)
CH2	72.15 (72.00)	4.06 (4.00)	11.14 (11.20)	4.02 (s, 2H, CH$_2$); 7.38 (s, 8H, ar)	250 (M$^+$, 98); 208 (M$^+$—OCN, 100)
E	66.59 (66.67)	3.12 (3.17)	11.02 (11.11)	7.14–7.51 (m, 8H, ar)	252 (M+, 100)
BPA	73.31 (73.38)	5.09 (5.04)	10.15 (10.07)	1.63 (s, 6H, CH$_3$); 7.35 (S, 8H, ar)	278 (M$^+$, 22); 263 (M$^+$—CH$_3$, 100)
4M	71.77 (72.00)	4.21 (4.20)	11.04 (11.20)	2.30 (s, 12H, CH$_3$); 3.81 (s, 2H, CH$_2$); 7.10 (S, 4H, ar)	306 (M$^+$, 100); 291 (M$^+$—CH$_3$, 56); 264 (M$^+$—OCN, 84)

^{a} In DMSO-d6; ^{b} singlet; ^{c} multiplet; ^{d} aromatic.

grams are shown in Figure 3. The decomposition temperatures at a 5% weight loss ranged between 403–439°C in nitrogen. The thermal stability is higher for the BT resins than the BMI/DDM or DGEDN/DDM system. Polymer BT$_{PB}$, with a *p*-phenylene structure, had the highest decomposition temperature and anaerobic char yield.

Thermal stability of BT$_{4M}$ containing methyl substituents was slightly lower than BT$_{CH2}$, which has no substituent.[18] This may be attributed to the lower crosslink density of BT$_{4M}$ than

that of BT$_{CH2}$, and also to the lower dissociation energy of the C—C bond than that of the C—H bond (83 v.s. 99 kcal/mol). Similarly, the thermal stability of BT$_E$ containing ether linkage was less than that of BT$_{CH2}$, which is also attributed to the lower dissociation energy of the C—O bond compared to that of the C—H bond (86 v.s. 99 kcal/mol).

Dynamic Mechanical Properties of Polymers

The dynamic mechanical properties of the cured BT resins with various molar ratios of the BMI/dicyanate esters (BPA) were examined, and the
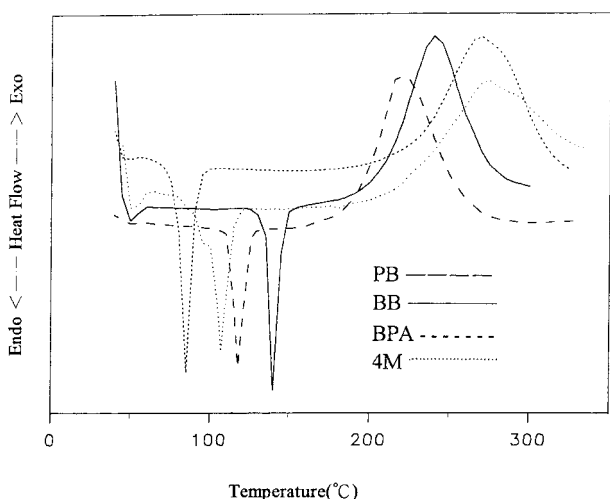

Figure 2 Representative DSC scans of dicyanate ester.

Table II   Thermal Analysis of Various Cyanate Ester Monomers

	T_m^{a} (°C)	T_i^{b} (°C)	$T_i - T_m$ (°C)	T_{exo}^{c} (°C)
PB	118	187	69	221
BB	141	205	64	240
CH2	113	208	95	260
E	92	203	111	258
BPA	83	216	133	268
4M	107	218	111	272

^{a} Melting point from d.s.c. with a heating rate of 20°C/min.
^{b} Onset temperature for curing reaction.
^{c} Maximum temperature of polymerization exotherm from d.s.c. with a heating rate of 20°C/min.

Table III   Thermal Analysis of Various Cyanate Ester with Bismaleimide Cured at 2/1 Molar Ratio with Catalyst

	T_i[a] (°C)	T_{exo}[b] (°C)	T_g[c] (°C)
BT_{pB}	125	165	322
BT_{BB}	143	190	305
BT_{CH2}	143	204	266
BT_E	137	194	255
BT_{BPA}	144	204	269
BT_{4M}	158	238	250

[a] Onset temperature for curing reaction.
[b] Maximum temperature of polymerization exotherm from d.s.c. with a heating rate of 20°C/min.
[c] Glass transition temperature from D.M.A.

results are shown in Figure 4. Both storage modulus and glass transition temperature decreased with the increase in dicyanate ester concentration. It may be attributed to the presence of aromatic ether linkage, which formed during cyclotrimerization, had increased the flexibility of the cured network. On the other hand, the higher the bismaleimide content in the formulation, the higher the modulus of the cured BT resin. The glass temperatures increased gradually with the increase in bismaleimide concentration initially; however, the increase became drastic when the BMI/dicyanate ratio exceeded 1 : 1, as shown in Figure 5.

The DMA polts of the storage moduli as a function of temperature for various cyanate monomers in BT resins are shown in Figure 6. The peak of tan δ was identified as the glass transition tem-

Table IV   Thermogravimetric Analysis of BMI/ Cyanate Esters Cured at Various Molar Ratio[c]

	$T_{5\%}$[a] (°C)	$T_{10\%}$[a] (°C)	T_{max}[b] (°C)	Char Yield at 600°C (%)
$BT_{1/0}$	475	487	491	67
$BT_{4/1}$	431	449	434	71
$BT_{2/1}$	434	443	434	64
$BT_{1/1}$	429	435	440	47
$BT_{1/2}$	425	433	434	62
$BT_{1/4}$	413	429	427	62
$BT_{0/1}$	401	423	428	42

[a] Temperature at which 5 or 10% weight loss was recorded by means of thermogravimetry at a heating rate of 20°C/min.
[b] Maximum polymer decomposition temperature.
[c] Cured at 180°C for 2 h and 250°C for 4 h.

Table V   Thermogravimetric Analysis of Various Cyanate Esters with Bismaleimide Cured at 2/1 Molar Ratio[c]

	$T_{5\%}$[a] (°C)	$T_{10\%}$[a] (°C)	T_{max}[b] (°C)	Char Yield at 600°C (%)
BT_{PB}	439	461	464	70
BT_{BB}	433	441	458	68
BT_{CH2}	431	441	445	64
BT_E	418	429	432	60
BT_{BPA}	425	433	434	62
BT_{4M}	403	423	435	53
BMPN/DDM[d]	400	429	492	61
BMPM/DDM[e]	395	419	447	56
DGEDN/DDM[f]	349	371	403	48

[a] Temperature at which 5 or 10% weight loss was recorded by means of thermogravimetry at a heating rate of 20°C/min.
[b] Maximum polymer decomposition temperature.
[c] Cured at 180°C for 2 h and 250°C for 4 h.
[d] BMPN cured with 4,4-diaminodiphenylmethane (DDM).
[e] Bis(4-maleimidophenyl)methane(BMPM) cured with DDM.
[f] Diglycidyl ether of 2,7-dihydroxynaphthalene cured with DDM.

peratures (T_g) and are listed in Table III. Polymer BT_{PB} and BT_{BB} containing p-phenylene and biphenylene exhibited a higher storage moduli and glass transition temperatures than the rest of the BT resins.

Moisture Resistance

The absorbed moisture acts as plasticizer, which reduces the mechanical and thermal properties of cured resins and also has a detrimental effect on
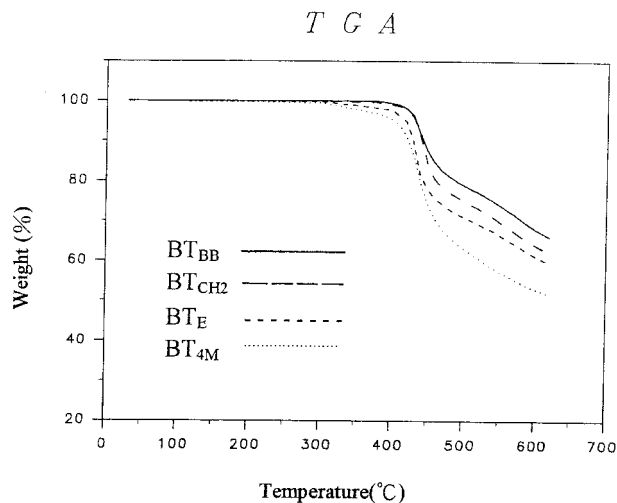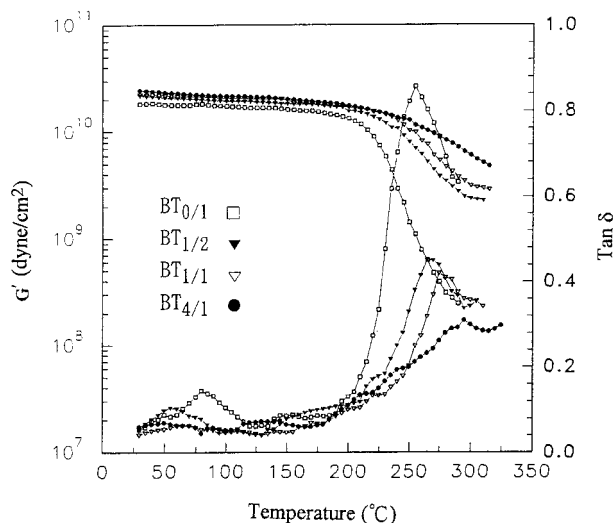

Figure 3   TGA curves of some cured BT resin.

Figure 4 DMA results of BT resins with various mol ratios.

Figure 6 DMA results of BT resins with various cyanate ester structures.

dielectric properties. The weight gains of cured polymers through 72-h exposure in boiling water ranged from 0.7–1.6%. The dielectric constant for a series of BT resins and comparable thermoset resins were measured at 30°C and $1M$ Hz in their dry state. BT resins had a lower dielectric constant and absorbed much less water than the BMI/DDM or DGEDN/DDM systems, as shown in Figure 7. The results are attributed to the relatively weak dipoles of the triazine structure.[17] The cured BT$_{4M}$ resin absorbed the least moisture, and
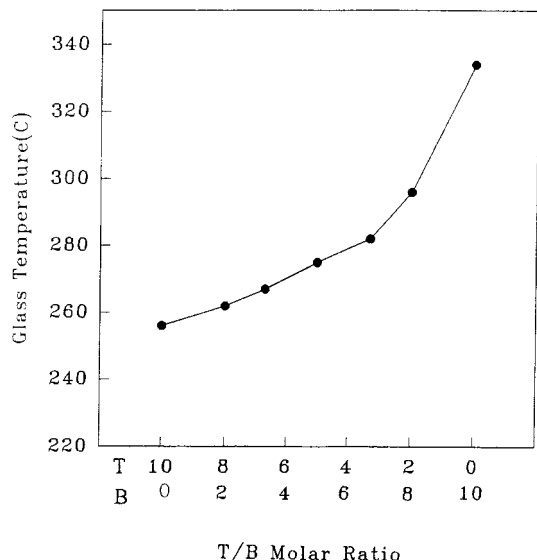
it is explained by the hydrophobic nature of its methyl substituents.

CONCLUSION

A series of naphthalene containing novel bismaleimide · triazine resins were prepared from various dicyanate ester and a bismaleimide (BMPN). The structure of the dicyanate ester controlled the reactivity of the polymerization. Cured BT resin with higher BMPN content had a higher glass transition temperature, thermal stability, and modulus. The water absorptions of bismaleimide · triazine resins were lower than that of the BMPN cured with the DDM system. All BT resins

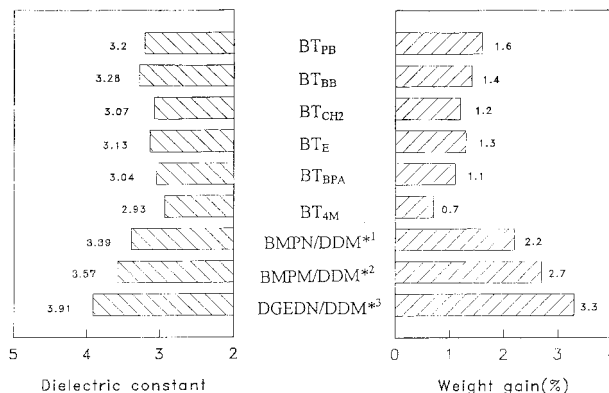**Figure 5** Relationship between B (BMI)/T (cyanate ester) mol ratio and T_g.

Figure 7 Dielectric constant and moisture absorption of various BMI copolymers.

had good heat-resistant and moisture-resistant properties. Thus, the naphthalene containing bis-maleimide·triazine resins are considered to be candidates for processable high-performance polymeric materials.

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