

Preparation and Characterization of Siliconized Epoxy-1,2-bis(maleimido)ethane Intercrosslinked Matrix Materials

A. Ashok Kumar, K. Dinakaran, M. Alagar

Department of Chemical Engineering, Anna University, Chennai 600 025, India

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ABSTRACT: Novel intercrosslinked networks of siliconized epoxy-1,2-bis(maleimido)ethane matrix systems are developed. The siliconization of epoxy resin is carried out by using 5–15% hydroxyl-terminated poly(dimethylsiloxane) with γ -aminopropyltriethoxysilane as a crosslinking agent and dibutyltin dilaurate as a catalyst. The siliconized epoxy systems are further modified with 5–15% 1,2-bis(maleimido)ethane and cured by using diaminodiphenylmethane. The prepared neat resin castings are characterized for their mechanical properties. Mechanical studies indicate that the introduction of siloxane into these epoxy resins improves the toughness with a reduction in the stress–strain values, whereas incorporation of bismaleimide (BMI) into the epoxy resin improves the stress–strain properties with a lowering of the toughness. The introduction of both siloxane and BMI into the epoxy resin influences the mechanical properties

according to their content percentages. Differential scanning calorimetry (DSC), thermogravimetry, and heat distortion temperature analyses are also carried out to assess the thermal behavior of the matrix materials that are developed. DSC thermograms of the BMI modified epoxy systems show unimodal reaction exotherms. The glass-transition temperature, thermal degradation temperature, and heat distortion temperature of the cured BMI modified epoxy and siliconized epoxy systems increase with increasing BMI content. The water absorption behavior of the matrix materials is also studied. © 2003 Wiley Periodicals, Inc. *J Appl Polym Sci* 89: 3808–3817, 2003

Key words: epoxy resin; bismaleimide; hydroxyl-terminated poly(dimethylsiloxane); siliconized epoxy; glass-transition temperature; plain strain fracture toughness

INTRODUCTION

The modern scenario of the development of new polymer-based engineering materials mainly depends on the modification of existing polymeric materials. At present, there is an increasing need for the development of polymeric materials with light weight, high fracture toughness, better impact strength, good thermal stability, and improved mechanical properties in order to utilize them for high performance engineering and aerospace applications. Epoxy resins are widely used as advanced materials, especially in the aerospace and electronic industries, because of their valuable physicochemical, thermal, mechanical, and electrical properties. However, their drawbacks, such as brittleness, moisture absorption, and a relatively low glass-transition temperature, limit their utility in some advanced high performance applications. Hence, the development of a suitable polymeric toughening mechanism is essential in order to improve the impact resistance and enhance the strain to

fracture, weather resistance, and glass-transition temperature of epoxy resins.

Many efforts have been made to improve the toughness of cured epoxy resins. Most of the earlier studies on the modification of epoxy resins were carried out by using carboxy-, amine-, and hydroxyl-terminated butadiene-acrylonitrile rubbers; functionally terminated acrylates; poly(phenylene oxide); and alkylene oxides.^{1–9} However, these modifications only result in improvement in the impact behavior with a significant loss of thermal and mechanical properties. The most common strategy employed for a toughening process is the development of an intercrosslinked polymer network having flexible and rigid molecular segments. An intercrosslinking network mechanism provides matrix materials with a single glass-transition temperature as compared with polymer blends that normally reveal multiple glass transitions that are due to the thermodynamic incompatibility of the components involved. Hydroxyl-terminated poly(dimethylsiloxane) (HTPDMS) and bismaleimide (BMI) have been selected to provide flexible and rigid segments, respectively, to the epoxy system.

HTPDMS has often been chosen as a toughener for epoxy resins, because it possesses high thermal and thermooxidative stability, high moisture resistance, good dielectric properties, and UV and chemical resistance.^{10–18} In our earlier work we observed that

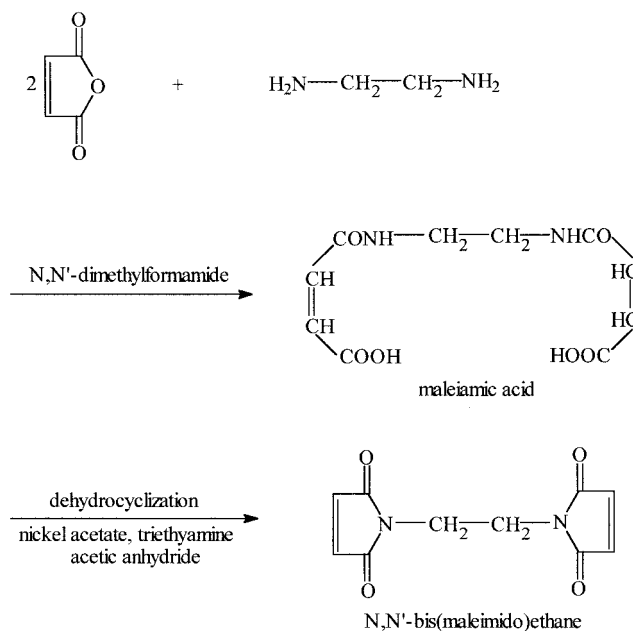
Correspondence to: M. Alagar (mkalagar@yahoo.com).

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siloxane incorporation into epoxy resins improves the impact strength and reduces the stress-strain properties with an insignificant change in the glass-transition behavior.¹⁹

In order to prevent the loss of stress-strain properties, the modification of siliconized epoxy systems with rigid materials like BMIs is essential because of the following superior thermomechanical properties: high crosslinking ability, high glass-transition temperature, high thermal stability, high char yield, excellent fire resistance, superior specific strength and specific modulus, and low water absorption.²⁰⁻²⁶

This work presents the preparation of siliconized epoxy-1,2-bis(maleimido)ethane intercrosslinked matrix materials by using HTPDMS and 1,2-bis(maleimido)ethane as chemical modifiers, γ -aminopropyltriethoxysilane (γ -APS) as a crosslinking agent, and dibutyltin dilaurate as a catalyst. In addition, we also investigate the mechanical, thermal, and water absorption properties of the siliconized epoxy-1,2-bis(maleimido)ethane intercrosslinked matrix materials.



Scheme 1 The synthesis of 1,2-bis(maleimido)ethane.

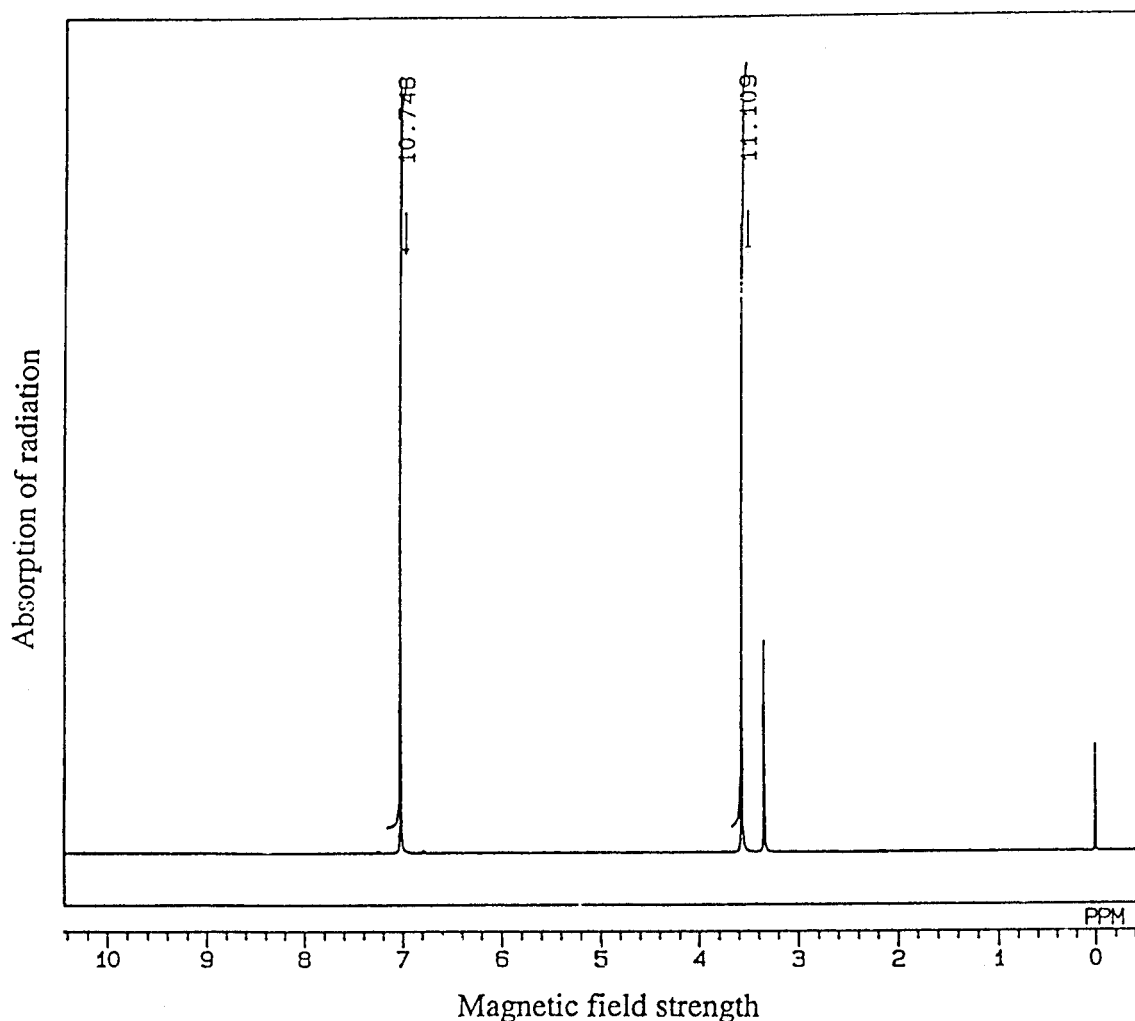


Figure 1 The ^1H -NMR spectra of 1,2-bis(maleimido)ethane.

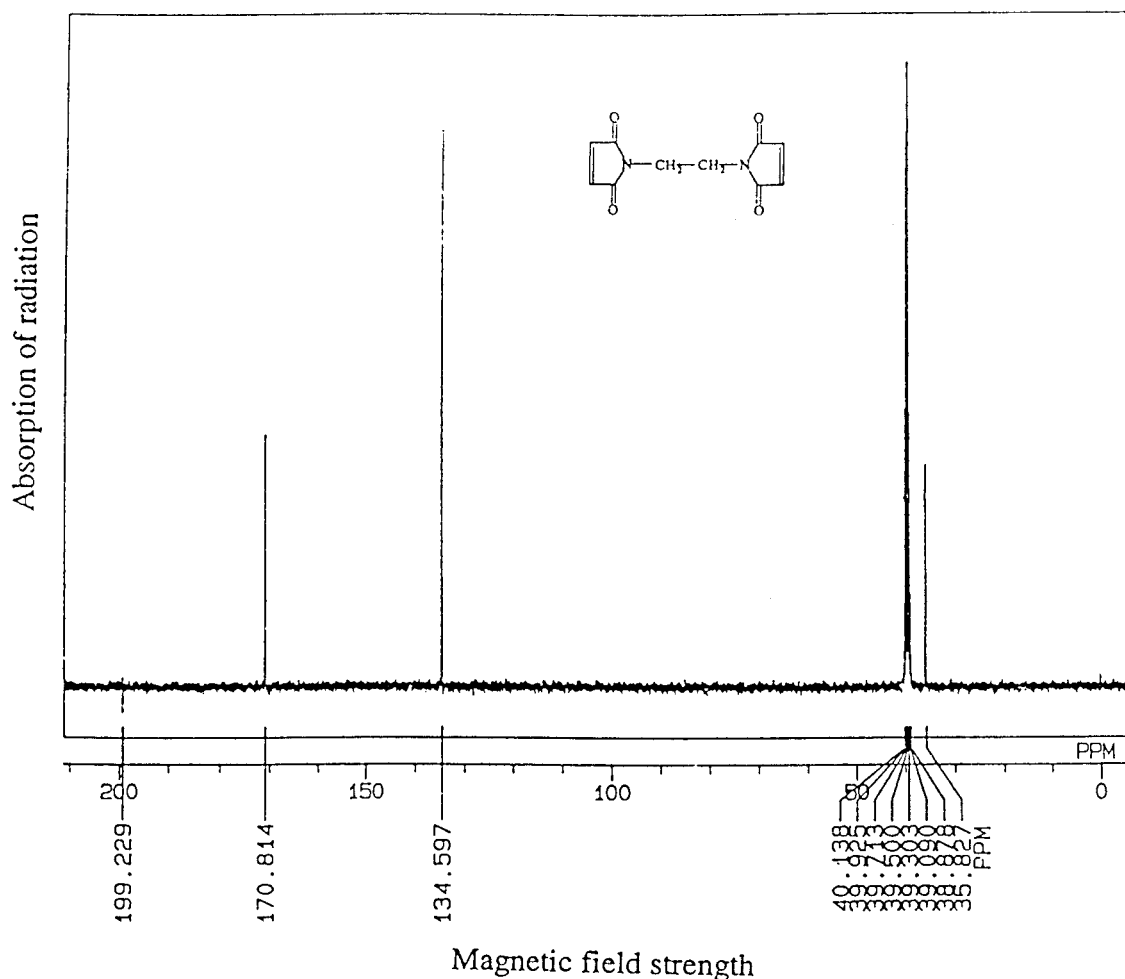


Figure 2 The ^{13}C -NMR spectra of 1,2-bis(maleimido)ethane.

EXPERIMENTAL

Materials

An epoxy resin (diglycidyl ether of bisphenol A, LY556; epoxy equivalent = 180–190, viscosity = $\sim 10,000$ cP) and 4,4-diaminodiphenylmethane (DDM) were obtained from Ciba-Geigy Ltd., India. 1,2-Bis(maleimido)ethane was synthesized from maleic anhydride and ethylenediamine. HTPDMS (molecular weight = 17,000–18,000, viscosity = 800–900 cP) was prepared from octamethylcyclotetrasiloxane by the ring-opening polymerization technique. The γ -APS crosslinking agent and dibutyltin dilaurate catalyst were obtained from Union Carbide and Merck, respectively.

Preparation of siliconized epoxy prepolymer

Fixed amounts of epoxy resin, varying amounts of HTPDMS, a stoichiometric amount of γ -APS (with respect to the hydroxyl groups of HTPDMS), and the dibutyltin dilaurate catalyst were thoroughly mixed at 90°C for 10 min with constant stirring. The product

was then degassed to remove the ethanol that was formed during the condensation reaction between γ -APS and HTPDMS. The reaction scheme is presented in an earlier communication.²¹

Preparation of 1,2-bis(maleimido)ethane

1,2-Bis(maleimido)ethane was prepared according to a slightly modified reported procedure²⁷ from ethylenediamine and maleic anhydride using acetic anhydride as the dehydrating agent with nickel acetate and triethylamine as catalysts, as shown in Scheme 1. To a 1-L three-necked flask fitted with a paddle stirrer, reflux condenser, and nitrogen inlet were added 300 mL of freshly distilled *N,N*-dimethyl formamide, 0.5 mol (49.0 g) of maleic anhydride, and 0.25 mol (98.1 g) of ethylenediamine. The temperature of the solution rose to 115°C and its opaque color changed to yellow. Stirring was thereafter continued for 30 min at 90°C . Then 0.5 g of nickel(II) acetate, 25 mL of triethylamine, and 118 mL of acetic anhydride were added to the reaction vessel while maintaining the system at 90°C for another 30 min. The reaction mixture was cooled to

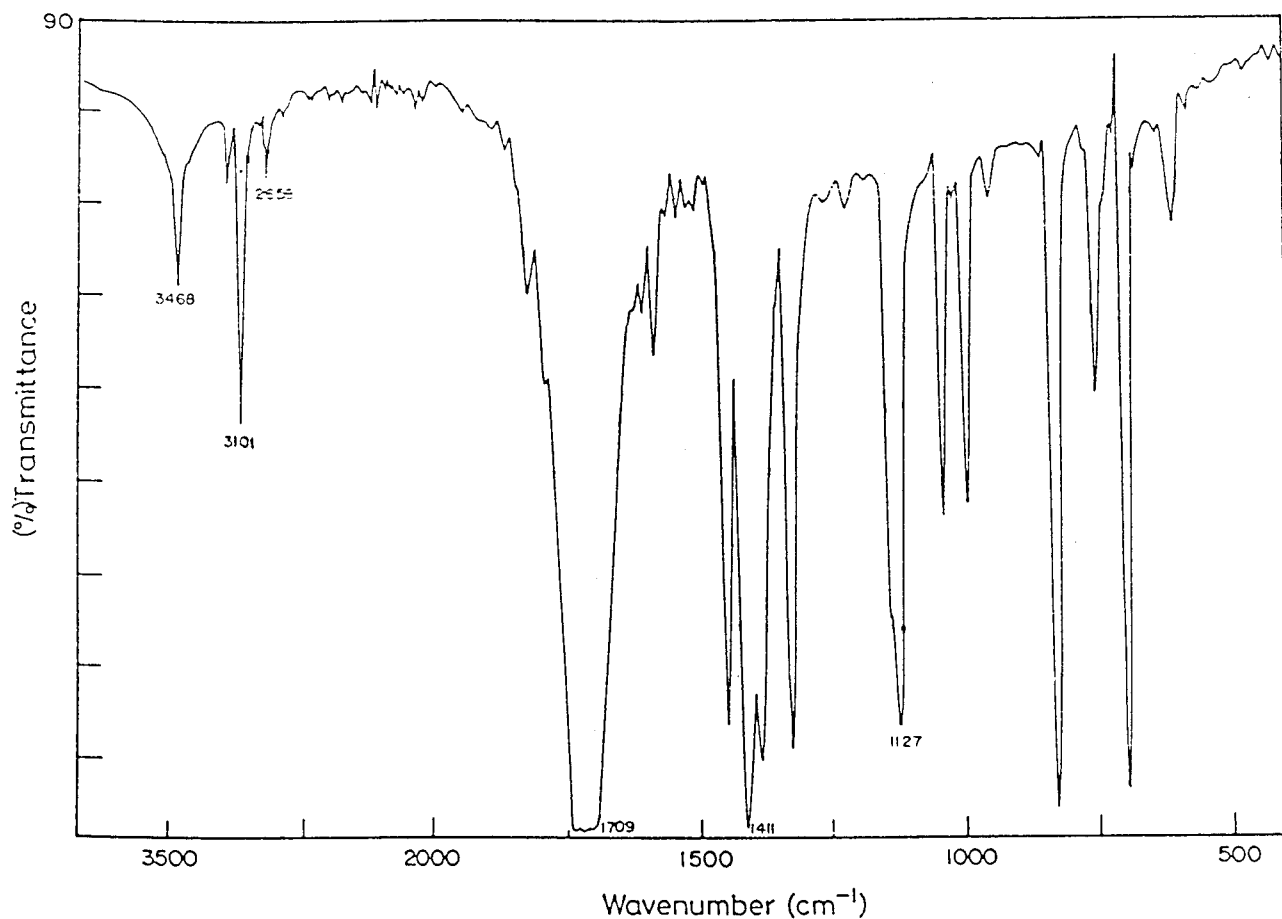


Figure 3 The FTIR spectra of 1,2-bis(maleimido)ethane.

TABLE I
Mechanical Properties of Epoxy, Siliconized Epoxy, BMI Modified Epoxy,
and BMI Modified Siliconized Epoxy Systems

Matrix system	Epoxy/ HTPDMS/BMI composition	Tensile strength (MPa)	Tensile modulus (MPa)	Flexural strength (MPa)	Flexural modulus (MPa)	Plain strain fracture toughness (MPa m ^{1/2})
A	100/00/00	63.2 ± 9	2751.7 ± 39	110.0 ± 10	1812.9 ± 42	1.14 ± 0.10
B	100/05/00	40.1 ± 3	2272.9 ± 30	70.6 ± 6	1735.4 ± 39	2.31 ± 0.05
C	100/10/00	33.6 ± 4	2119.9 ± 29	52.3 ± 4	1710.9 ± 28	2.61 ± 0.04
D	100/15/00	26.6 ± 5	1852.1 ± 40	49.1 ± 2	1373.4 ± 31	2.93 ± 0.15
E	100/05/05	43.4 ± 3	2366.2 ± 25	76.6 ± 3	1795.8 ± 30	1.35 ± 0.08
F	100/05/10	52.7 ± 3	2405.4 ± 28	85.2 ± 2	1882.6 ± 25	1.28 ± 0.11
G	100/05/15	58.6 ± 4	2472.1 ± 20	99.2 ± 4	1927.3 ± 32	1.20 ± 0.05
H	100/10/05	37.7 ± 4	2160.2 ± 35	61.8 ± 3	1756.0 ± 27	1.79 ± 0.11
I	100/10/10	41.9 ± 3	2371.4 ± 30	68.0 ± 3	1792.1 ± 35	1.62 ± 0.09
J	100/10/15	47.9 ± 5	2425.4 ± 31	71.8 ± 4	1825.2 ± 31	1.50 ± 0.05
K	100/15/05	30.2 ± 4	1891.9 ± 27	55.8 ± 3	1422.5 ± 30	1.97 ± 0.08
L	100/15/10	36.9 ± 4	2061.8 ± 31	64.1 ± 4	1532.2 ± 25	1.70 ± 0.05
M	100/15/15	41.1 ± 3	2166.2 ± 26	68.9 ± 3	1602.7 ± 27	1.54 ± 0.09
N	100/00/05	65.4 ± 5	2792.7 ± 30	114.8 ± 5	1891.8 ± 27	0.92 ± 0.07
O	100/00/10	68.1 ± 6	2843.3 ± 35	120.2 ± 4	2002.3 ± 33	0.85 ± 0.12
P	100/00/15	75.7 ± 4	2935.4 ± 37	126.2 ± 7	2196.2 ± 38	0.79 ± 0.07

^a HTPDMS, hydroxyl-terminated poly(dimethyl siloxane);

^b BMI, 1,2-bis(maleimido)ethane.

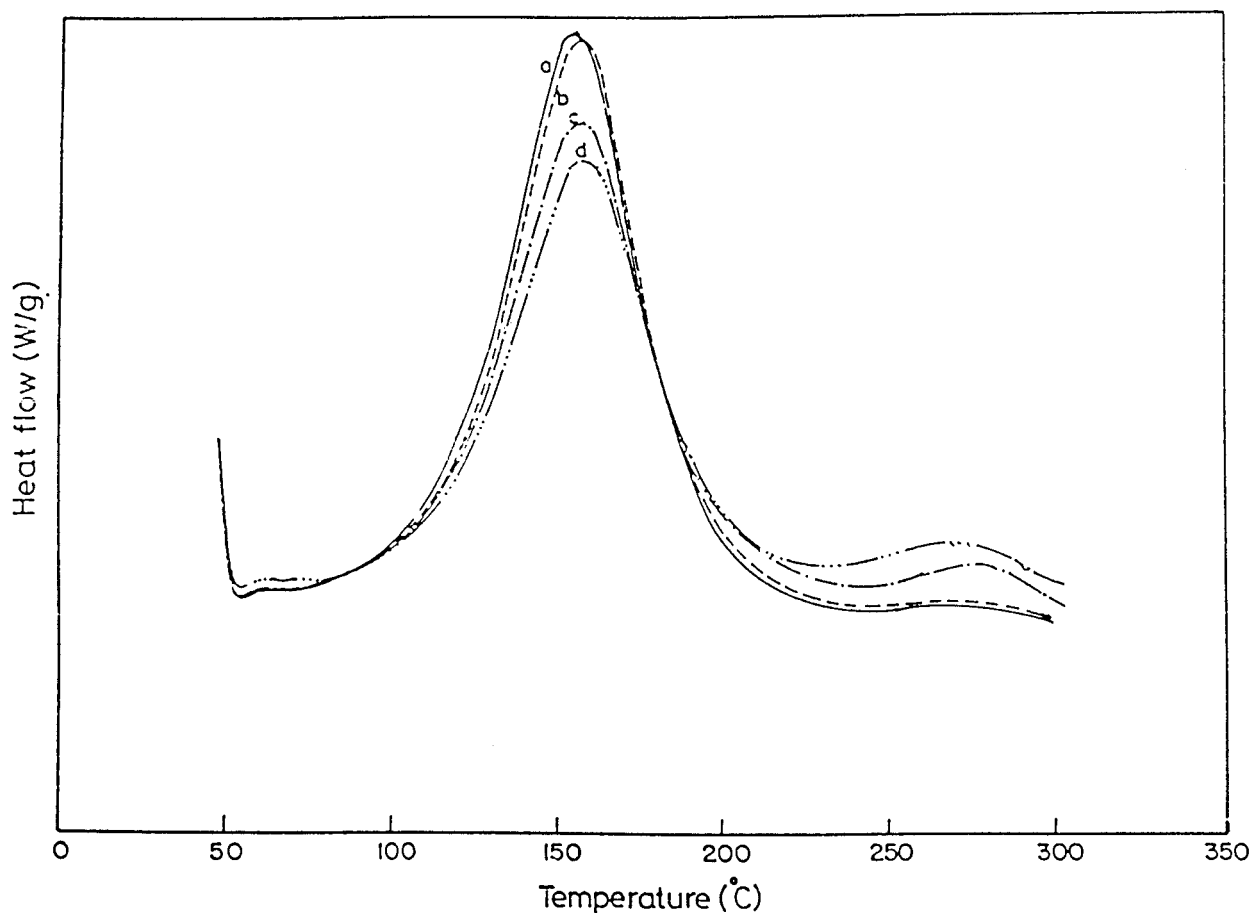


Figure 4 DSC thermograms of epoxy-BMI systems during cure: (a) epoxy-DDM, (b) epoxy-DDM-5% BMI, (c) epoxy-DDM-10% BMI, and (d) epoxy-DDM-15% BMI.

30°C, poured into 2 L of ice water, and stirred for 10 min to crystallize the BMI. The resulting brown precipitate was filtered off by suction, washed twice with 1 L of water, and filtered. For further purification, the product was dissolved in 300 mL of dry acetone, refluxed with 15 g of charcoal, and filtered. The volume of the solution was reduced to 100 mL by evaporation and then kept at 0°C overnight. The resulting precipitate was filtered off by suction and dried.

Preparation of siliconized epoxy BMI blend

The stoichiometric amount of DDM was dissolved in siliconized epoxy at 90°C with vigorous stirring. After complete dissolution, the calculated amount of 1,2-bis(maleimido)ethane was added. Agitation was continued at 110°C until a homogeneous product was obtained. The product was subjected to a vacuum to remove trapped air and then cast and cured at 120°C for 1 h. The castings were postcured at 150°C for 2 h and 200°C for 1 h and finally removed from the mold and characterized.

Test methods

The FTIR spectra were recorded using a Perkin-Elmer 781 IR spectrometer with KBr pellets. The ^1H - and ^{13}C -NMR spectra were recorded on a Bruker 400-MHz NMR spectrometer with dimethylsulfoxide- d_6 (DMSO- d_6) as the solvent.

The tensile (stress-strain) properties were determined with dogbone-shaped specimens according to ASTM D3039 using an Instron testing machine (model 6025) at a crosshead speed of 2 mm/min. The flexural strength was measured according to ASTM D790. The fracture toughness of the neat resin castings under conditions of plain strain was evaluated as per ASTM E399-74. As many as five test pieces were used to generate the data points for the mechanical tests.

The glass-transition temperature (T_g) of the samples was determined using a differential scanning calorimetry (DSC) 2910 instrument (TA Instruments) in the temperature range of 50–250°C at a heating rate of 10°C/min. Thermogravimetric analysis was carried out using a Thermal Analyst 2000 (TA Instruments) at a heating rate of 10°C/min in an inert atmosphere.

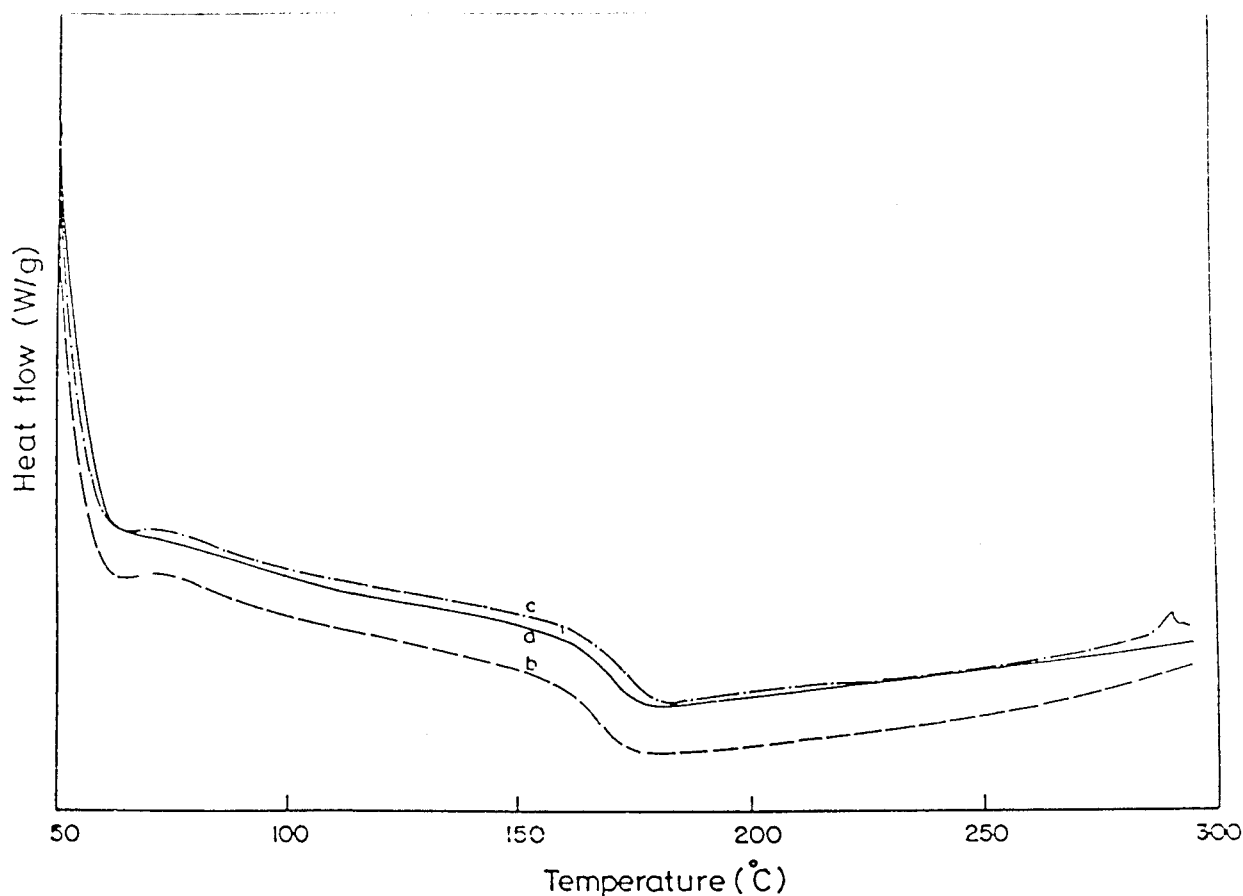


Figure 5 DSC traces of 10% siliconized epoxy systems with (a) 0, (b) 5, (c) 10, and (d) 15% BMI.

The heat deflection temperature (HDT) of the samples was tested as in ASTM D648-72. The HDT measures the temperature at which deformation occurs when the composite specimens (127-mm length, 13-mm thickness, 3-mm width) were placed in an oil bath under a load of 1.82 MPa, and the temperature was raised at a rate of 2°C/min. The temperature at which the specimen was deflected by 0.25 mm was noted. The deflection was monitored by a linear variable-displacement transducer.

The water absorption properties of each sample was tested according to ASTM D570. The cured specimens, (60-mm length, 3-mm thickness, 60-mm width) were immersed in distilled water for 24 h. Then the specimens were withdrawn and the surface water removed using tissue paper, and they were weighed to an accuracy of 0.001 g.

RESULTS AND DISCUSSION

Spectral analysis

The ^1H - and ^{13}C -NMR spectra of the synthesized BMI [1,2-bis(maleimido)ethane] are shown in Figures 1 and 2. ^1H -NMR (DMSO- d_6 , δ): 7.01 (s, 4H, olefinic); 3.56 [s, 4H, (N—CH $_2$) $_2$]. ^{13}C -NMR (DMSO- d_6 , δ): 35.8 (CH $_2$);

134.5 (C=C); 170.8 (C=O). Figure 3 presents the IR spectrum of 1,2-bis(maleimido)ethane. IR (KBr, ν): 3101 (=C—H); 2658 (C—H); 3468, 1709 (C=O); 1411, 1127 (C—N—C).

Mechanical properties

The mechanical properties of the cured epoxy resin, siliconized epoxy resin, BMI modified epoxy resin, and BMI modified siliconized epoxy resin systems are presented in Table I. Incorporation of HTPDMS into an epoxy resin decreases the tensile and flexural strength; this is due to the presence of flexible siloxane linkages having free rotation of the —Si—O—Si— bond, the weak interfacial attraction force of pendant methyl groups present in the HTPDMS, and the weak interfacial boundary between siloxane and the epoxy matrix. The incorporation of aliphatic BMI into the epoxy system increases the tensile and flexural strength because the incorporation of BMI increases the crosslink density and causes the formation of an intercrosslinking network. The increases in tensile strength for 5, 10, and 15% incorporation are 3.5, 7.7, and 19.7%, respectively, when compared with the unmodified epoxy system (system A). The increases in

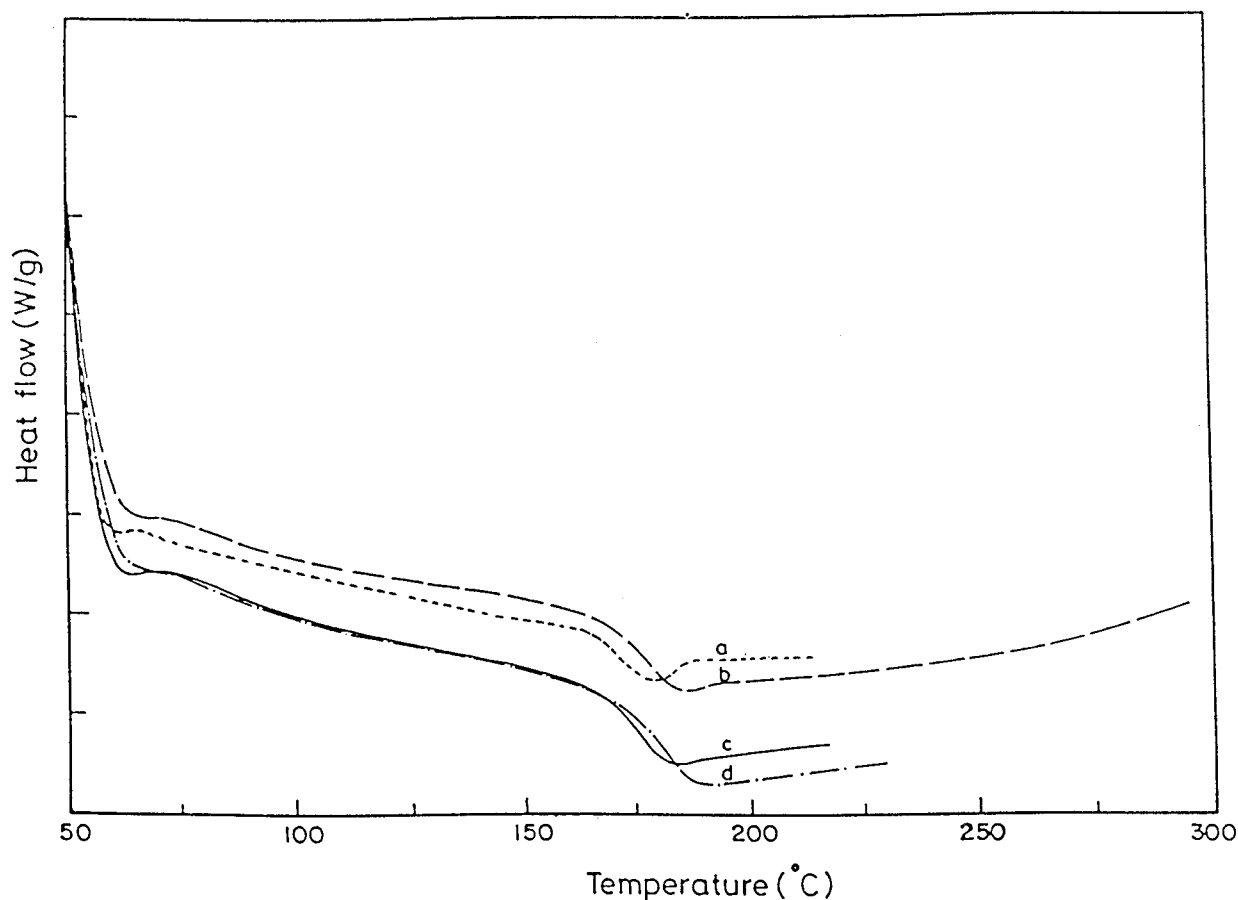


Figure 6 DSC traces of epoxy systems with (a) 0, (b) 5, (c) 10, and (d) 15% BMI.

flexural strength for 5, 10, and 15% incorporation are 4, 9.2, and 14.7%, respectively. The increases in both the tensile strength and flexural strength are slightly lower in the case of aliphatic BMI modified epoxy systems compared to the same percentage of incorporation of aromatic BMI (*N,N'*-bismaleimido-4,4-diphenylmethane) modified epoxy systems.²⁰⁻²² For example, the increases in the tensile strength and flexural strength for the 10% aromatic BMI modified epoxy system are 12.3 and 15.7%, respectively, whereas for the 10% aliphatic BMI modified epoxy system the increases are 7.7 and 9.2%, respectively. The greater improvement in the tensile and flexural strengths for the aromatic BMI modified epoxy system is attributable to the presence of a rigid aromatic ring. The brittle behavior of aliphatic BMI may also be one of the reasons for less improvement in the tensile and flexural properties in the aliphatic BMI modified epoxy systems.

The introduction of combinations of both siloxane and BMI into epoxy resins (systems E-M) alters the tensile strength and flexural strength according to their content percentage. The values of the tensile and flexural strengths of the siliconized epoxy system steadily increase with increasing BMI content. The

highest tensile and flexural strength values of 58.6 and 99.2 MPa are obtained for the epoxy system modified with the combination of 5% siloxane and 15% BMI (system G). Similarly, the lowest tensile and flexural values of 30 and 55.8 MPa are obtained for the epoxy system modified with 15% siloxane and 5% BMI (system K). The other systems exhibit tensile and flexural behavior between the above two extremes. The values of the tensile modulus and flexural modulus exhibit similar trends to those observed in the tensile and flexural strengths.

The incorporation of siloxane into an epoxy resin enhances the toughness according to the percentage of siloxane content, which is due to high-energy absorption and the resilient behavior of the flexible siloxane molecule. The incorporation of BMIs in both epoxy and siliconized epoxy decreases the toughness, and this is due to an increase in the crosslinking density. Incorporation of 5, 10, and 15% BMI into the epoxy resin decreases the toughness behavior by 19.2, 25.4, and 30.7%, respectively. The decrease in toughness is higher in the aliphatic BMI modified epoxy systems compared to those of aromatic BMI modified epoxy systems. For example, the decrease in toughness for the 10% aliphatic BMI modified epoxy system is 25.4%

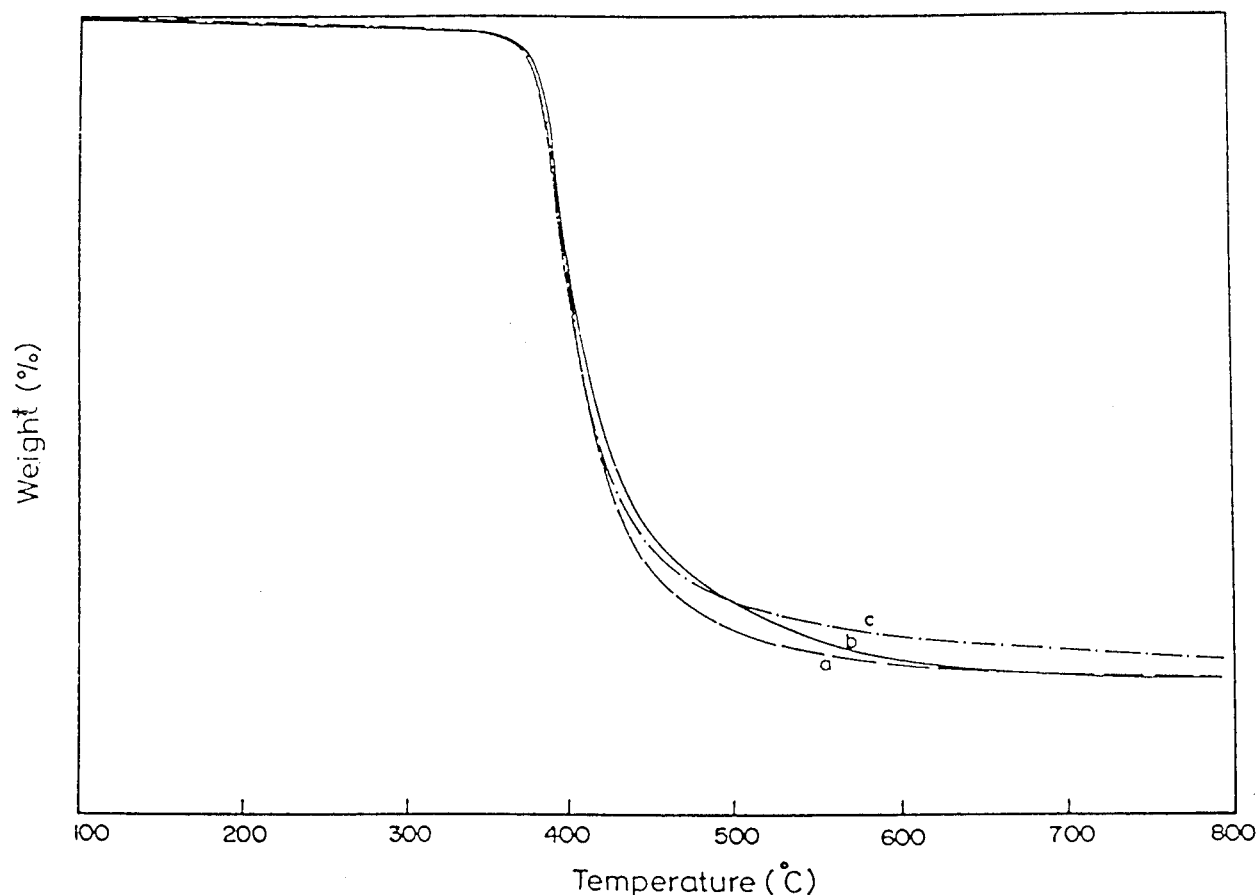


Figure 7 Thermogravimetric analysis curves of 10% siliconized epoxy systems with (a) 0, (b) 5, (c) 10, and (d) 15% BMI.

whereas for aromatic BMI modified systems it is 8.7%. This is attributable to a higher number of reactive molecules being present in the aliphatic BMI modified systems compared to that for the same percentage of aromatic BMI modified epoxy systems, which in turn enhances the crosslink density. Moreover, the available free volume for molecular relaxation in the aliphatic BMI is less than that of aromatic BMI modified systems, because of the linear molecular structure, which in turn favors close, rigid packing and thus reduces the molecular relaxation. In the aromatic BMI modified systems, the complex shapes prevent close packing coupled with high energy of absorption of the aromatic ring structure and more free volume is available for molecular relaxation.

Thermal properties

Figure 4 presents the DSC thermograms of the epoxy resin containing 5, 10, and 15% aliphatic BMI. Similar to the aromatic BMI modified epoxy systems,²¹ the aliphatic BMI incorporated epoxy systems show unimodal reaction exotherms that are due to a lower concentration. For the aliphatic BMI incorporated epoxy systems, the reaction commences at 120°C and

reaches peak maxima of 175, 177, and 180°C for 5, 10, and 15%, respectively. The large exothermic peaks for the aliphatic BMI incorporated systems are due to the following reactions: an oxirane ring-opening reaction with the active amine hydrogens of DDM, an autocatalytic reaction of the oxirane ring with pendant hydroxyl groups of the epoxy resin and hydroxyl groups formed during the reaction, an addition reaction of the $-\text{NH}_2$ groups of DDM with the double bonds of BMI (Michael addition), and a BMI homopolymerization reaction.

The glass-transition temperatures (T_g) of the matrix systems are presented in Figures 5 and 6. In our earlier work we observed that incorporation of HTPDMS into an epoxy resin has no significant effect on the T_g . However, a slight decreasing trend in the T_g was noticed with an increasing HTPDMS concentration.²¹ The incorporation of aliphatic BMI into both epoxy and siliconized epoxy systems enhances the T_g values. This may be explained as being due to the formation of an intercrosslinking network between the epoxy or siliconized epoxy and BMI. The values of the T_g for 5, 10, and 15% BMI modified epoxy systems are 178, 188, and 191°C, respectively. The single T_g value obtained

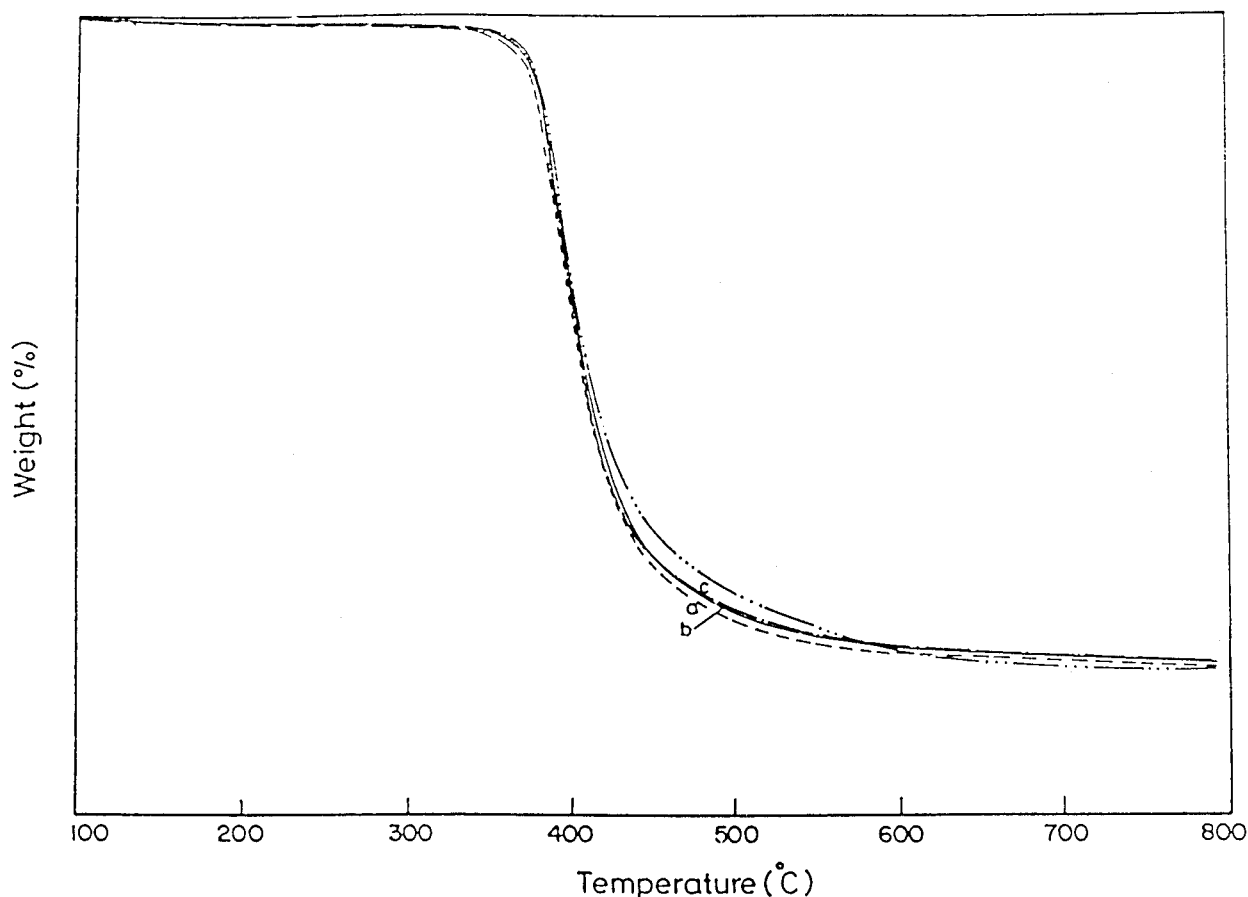


Figure 8 Thermogravimetric analysis curves of epoxy systems with (a) 0, (b) 5, (c) 10, and (d) 15% BMI.

for the BMI modified systems further confirms the presence of an intercrosslinked network structure.

Figures 7 and 8 show that the thermal degradation temperatures of both BMI modified epoxy resin and BMI modified siliconized epoxy resin systems are increased with increasing BMI content. The delay in degradation is mainly due to the thermally stable heterocyclic structure of BMI and the formation of an intercrosslinking network. The temperatures required for 10, 30, and 50% weight losses of the unmodified epoxy-DDM system are 365, 383, and 392°C, respectively, whereas the temperatures required to attain the same weight losses for the 10% BMI modified system are found to be increased to 391, 421, and 435°C, respectively. A similar trend is observed for 5 and 15% BMI modified epoxy systems and BMI modified siliconized epoxy systems. The thermal stability of the BMI modified siliconized epoxy system is slightly higher than that of BMI modified epoxy and HTPDMS modified epoxy systems. For example, the temperatures required for 50% weight loss of epoxy modified with 10% HTPDMS and epoxy modified with 10% BMI are 415 and 435°C, respectively; to attain the same weight loss (50%) for the epoxy system modified with 10% BMI and 10% HTPDMS, this increases to 440°C.

This enhancement is mainly due to the synergistic effect of BMI and HTPDMS.

The HDT values for epoxy, HTPDMS modified epoxy, BMI modified epoxy, and BMI modified siliconized epoxy systems are presented in Table II. From the table it is evident that the HDT is decreased with increasing HTPDMS concentration. However, an increasing trend in the HDT values is observed for BMI modified epoxy resin and BMI modified siliconized

TABLE II
Heat Deflection Temperature (HDT) and Water Absorption Properties of BMI-HTPDMS-Epoxy Matrices

Matrix system	Epoxy/HTPDMS/BMI composition	HDT	Water absorption (%)
A	100/00/00	153	0.1201
B	100/05/00	150	0.1152
C	100/10/00	145	0.1105
D	100/15/00	139	0.1032
H	100/10/05	142	0.0634
I	100/10/10	145	0.0546
J	100/10/15	149	0.0511
N	100/00/05	154	0.0778
O	100/00/10	155	0.0742
P	100/00/15	158	0.0611

epoxy resin systems. The enhancement in the HDT values for BMI modified systems is attributable to the high crosslinking density and rigid heterocyclic structure imparted by BMI.

Water absorption

The water absorption characteristics of the pure epoxy system, siliconized epoxy systems, and BMI modified epoxy and siliconized epoxy systems are presented in Table II. The water absorption test was carried out by immersing the specimen with appropriate dimensions in distilled water at 30°C for 24 h.

The incorporation of siloxane into the epoxy system decreases the water absorption behavior when its concentration is increased. For example, the water uptake for unmodified epoxy systems is 0.1201% whereas for 5, 10, and 15% siloxane incorporation the water uptake is 0.1152, 0.1105, and 0.1032%, respectively. The decrease in the percentage of water uptake for siloxane incorporated systems is due to the inherent hydrophobic nature of —Si—O—Si— linkage. The inclusion of BMI into epoxy and siliconized epoxy systems hardly affects the percentage of water uptake. This may be due to the fact that a more effective network is formed when increasing the BMI concentration, which in turn increases the crosslinking density and rigidity.

CONCLUSION

Siliconized epoxy/1,2-bis(maleimido)ethane intercrosslinked matrix systems were successfully developed. The mechanical and thermal properties were increased with an increasing percentage of aliphatic BMI content, as observed in the case of aromatic BMI incorporated systems. However, the values of the plain-strain fracture toughness were decreased with increasing aliphatic BMI content and the decrease in its value was higher in the aliphatic BMI modified epoxy systems compared to aromatic BMI modified systems. The single glass-transition temperatures obtained for the BMI modified epoxy and BMI modified siliconized epoxy systems clearly confirm the presence of an intercrosslinking network. The water absorption behavior of these epoxy systems decreases with the incorporation of aliphatic BMI and HTPDMS.

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