

Effects of Bismaleimide Resin on Dielectric and Dynamic Mechanical Properties of Epoxy-Based Laminates

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ABSTRACT

Effects of bismaleimide (BMI) resin on dielectric properties e.g., permittivity (ϵ'), loss factor (ϵ''), and ionic conductivity (σ) of two different epoxy-amine resin systems were measured during dynamic curing (over a temperature range 20-300°C) by using dielectric analysis (DEA). Dynamic mechanical properties, e.g., flexural storage and loss moduli (E' and E'') and shear storage and loss moduli (G' and G'') of the glass-fabric reinforced composites (laminates) of the above mentioned resin matrices were carried out by dynamic mechanical analysis (DMA) technique. Double exothermic curing peaks were observed for the formulations with DDS as curing agent. For each system, that region of the curve in which the values for ionic conductivity are frequency dependent, indicates that the measurement is strongly influenced by dipole relaxations. At temperatures near or below T_g , dielectric properties of resins are dominated by the molecular dipoles (frequency-dependent response). Ionic conductivity increases largely on using DDS as curing agent for DGEBA over MDA however; addition of BMI to DDS- and MDA-cured thermosets decreases the ionic conductivity in the former and increases it in the later resin systems. Addition of bismaleimide (BMI) resin in three different concentrations to the glass-fabric DGEBA/MDA and DGEBA/DDS laminates, increases T_g , $T_{g\infty}$, and flexural and shear storage moduli (E' and G') considerably.

Key Words:

epoxy-amine;
bismaleimide;
laminates;
DEA;
DMA.

INTRODUCTION

Thermosetting polyimides have several key properties, which lead them successfully in specialized applications. Typically, thermosetting polyimides circuit boards are used because of their inherent chemical resistance, heat stability

and overall reliability. The PCB-industry however, is still searching for some new resins with high glass transition temperature. The T_g ideally should be well above the soldering temperature, thus providing a low coefficient of

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thermal expansion. Besides property improvements, new candidates should also have all the processing characteristics of an epoxy resin.

Epoxy resins are widely used as adhesives and matrix materials for high performance composites. Linear epoxy resins are converted into a three-dimensional cross-linked thermoset network during curing. Various chemical reactions that take place during the curing determine the resin morphology which, in turn, it determines the physical and mechanical properties of the cured formulation.

It is easy to understand why attempts have been made to merge bismaleimide and epoxy resin technologies in the hope of achieving both the temperature performance of the polyimides and the processing ease of epoxy resins [1-5].

The objective of this work was to develop resin networks with improved dielectric, mechanical, and thermal properties. Therefore, bismaleimide (BMI)/aromatic diamine prepolymers were used to cure epoxy resin diglycidyl ether of bisphenol-A (DGEBA), in order to achieve the both above mentioned improvements.

DGEBA-based thermosets have been investigated using a stretched exponential or Kohlrausch-Williams-Watt relaxation function to describe the data [6,7]. The dielectric properties of a thermoset during its curing process are determined by its changing chemical composition as a result of chemical reactions, which decrease the concentration of certain molecular segments and increase the concentration of others. This change in chemical composition also causes a decrease in the diffusivity or an increase in the viscosity, which in turn decreases the probability of the occurrence of further chemical reactions [8,9]. The autocatalytic reactions thus become self-retarding as the increase in the viscosities increases the thermoset's T_g toward the value of the curing temperature (T_{cure}). At high viscosities as encountered near T_g , the rate of the chemical changes is controlled by the rate of molecular diffusion, rather than the law of mass action. Although at relatively lower viscosities, the latter one has been regarded as the controlling factor. From a physical point of view, cross-linking, that ultimately forms a network sterically hindered the translational and rotational diffusion of individual molecules in to the thermoset structure. As

in the case of linear-chain polymerization, the changes that occur during the densification of the network as a result of increased cross-linking are: (a) decreases in the mobility of the impure ions [10], and in the concentration and mobility of protons which may be produced by partial dissociation of amine and the maleimide groups, (b) a decrease in the contribution of the static dielectric permittivity per unit molecule as a result of increased chain length, and (c) a general decrease in the dipolar reorientation rates caused by an increased steric hindrance [7].

EXPERIMENTAL

Materials

The epoxy resin, diglycidyl ether of bisphenol-A (DGEBA (LY-556)), 4,4'-methylene dianiline (MDA, Fluka) and 4,4'-diamino diphenyl sulphone (DDS, Fluka) were used as receiveol.

4,4'-Bismaleimido diphenyl methane (BMI) was synthesized from the reaction of MDA with maleic anhydride using acetone as solvent [11,12].

Epoxy compatible E-glass fabric (woven mat, 360 g/m³) obtained from PMP Company, Iran was used for making the laminates (nine ply laminates). The resin systems studied are given in Table 1.

Dielectric Analysis

Dielectric analysis (DEA) was carried out using DuPont 2970 dielectric analyzer module which consists of a sensor and ram/furnace assembly,

Table 1. Compositions of the resin systems studied.

Sample No.	Resin System	Curing agent conc. (phr)	BMI conc. (wt %)
1	DGEBA/MDA	30.43	0
2	DGEBA/MDA/BM	30.43	5
3	DGEBA/MDA/BM	30.43	15
4	DGEBA/MDA/BM	30.43	25
5	DGEBA/DDS	36.34	0
6	DGEBA/DDS/BM	36.34	5
7	DGEBA/DDS/BM	36.34	15
8	DGEBA/DDS/BM	36.34	25

incorporated in a cabinet which contains the supporting electronics.

The experiments were performed in ceramic single surface mode since it is the optimum sensor arrangement for monitoring the cure reaction of thermosetting materials. These sensors contain an interdigitated array of excitation and response electrodes on a planar surface. A platinum resistance thermometer is contained on the surface of the sensor thus, providing accurate sample temperature measurements. Each sensor was calibrated before performing the experiment.

The classic Debye equations for permittivity, ϵ' , and loss factor, ϵ'' , are as follows:

$$\epsilon' = \epsilon_u + \frac{(\epsilon_r - \epsilon_u)}{1 + (2\pi f\tau)^2}$$

Where, first and second terms are the permittivity due to alignment of dipoles and induced dipoles, respectively and,

$$\epsilon'' = \frac{(\epsilon_r - \epsilon_u)2\pi f\tau}{1 + (2\pi f\tau)^2} + \frac{\sigma}{2\pi f\epsilon_0}$$

Where, first and second terms are dipole loss factor and ionic conductivity, respectively.

In these equations ϵ_u , ϵ_r , τ , and σ are unrelaxed permittivity, the relaxed permittivity, the molecular relaxation time, and the bulk ionic conductivity, respectively. The bulk ionic conductivity, σ , can be used to follow the rheological changes that take place during the curing of thermosets:

$$\sigma = \epsilon''\omega\epsilon_0$$

Where, σ , ω , f , and ϵ_0 are the bulk ionic conductivity, the angular frequency ($2\pi f$), the frequency (Hz), and the absolute permittivity of free space (8.85×10^{-12} F/m), respectively.

The blends (Table 1) were prepared by heating DGEBA to 60°C and adding the other component/components with continuous stirring till a homogeneous and clear solution was obtained (<5 min). Each freshly prepared sample was cooled to room temperature and then painted on to the calibrated DEA sensor using a wooden rod and heated from room temperature to 200-300°C (depending on the sample) at a rate of 5°C/min under nitrogen atmosphere (0.5-1 min). The dielectric properties were

measured at 5 different frequencies, i.e., 1×10^4 , 3×10^4 , 1×10^5 , 3×10^5 , and 1×10^6 Hz.

Preparation of Laminates

Nine pieces of the size 4" × 1.5" of epoxy compatible E-glass woven mat were weighed and coated with a 20% methyl ethyl ketone (MEK) solution of each of the resin systems given in Table 1. The prepregs were dried in an air oven at 110-120°C for 15-20 min. The dried prepregs were weighed again and stacked together in the mould and then placed between two steel plates, already sprayed with silicone mould release agent on the contact surface. The assembly was then placed in a hydraulic press, where the temperature increased gradually from room temperature to $170 \pm 5^\circ\text{C}$, to allow the resin to flow and thus, get uniformed. For all the systems the temperature was maintained at $170 \pm 5^\circ\text{C}$ for 1 h at a pressure of 20 tons/inch², which was applied gradually during the first 15 min. No difficulty was faced regarding the overflowing of resin from the mould due to gradual applying of heat and pressure. Thus, the obtained laminates were removed from the press after cooling and then kept for post-curing at 200°C in an air oven for several hours. The resin content of each laminate was obtained by TGA.

Dynamic Mechanical Analysis

The concept of DMA is based on observing the viscoelastic response of materials subjected to a small oscillatory strain.

The method of sinusoidal excitation and response is very useful for determination of the dynamic mechanical properties of polymeric materials [13]. The applied force and the resulting deformation, both vary sinusoidal with time; the rate is usually specified by the frequency f in Hz or $\omega = 2\pi f$ in rad/s. For linear viscoelastic behaviour, the strain alternates sinusoidal but is out of phase with respect to the stress. This phase lag results from the time necessary for molecular rearrangements and is associated with relaxation phenomena.

The DuPont 983 dynamic mechanical analyzer together with DuPont 9900 thermal analyzer were employed for present investigations. The laminates were cut into the following dimensions; Length: 20-22 mm, Width: 10-11 mm, and Thickness: 2.0-2.4 mm

(according to the requirement of the DMA instrument).

The experiments were performed in resonant frequency mode where each sample was heated from room temperature to 200-300°C (depending on the sample) at the heating rate of 5°C/min. Then the samples were allowed to cool slowly to room temperature in the drive assembly enclosed by the oven, and they were again heated at the same rate and in the same atmosphere. This process was repeated for each sample a number of times (3-5 times) till the infinite glass transition temperature ($T_{g\infty}$), was obtained from the tan delta curves.

RESULT AND DISCUSSION

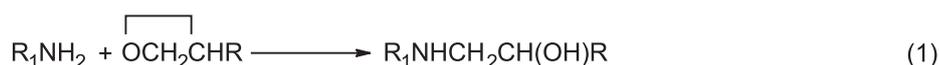
It is generally understood that, during the curing of these thermosets, the epoxide group combines with the primary amine ($-\text{NH}_2$) group of the cross-linking agent to form an $-\text{OH}$ group and secondary amine ($-\text{NHR}'$) group, which in turn it combines with another epoxide group to form an $-\text{OH}$ and ($-\text{NR}'\text{R}''$) group [14] as is shown in Scheme I.

In the Epoxy:Amine/BMI systems, besides the usual epoxide-amine reactions (reactions 1 and 2 and reaction 3, in some cases, in scheme I) there are three primary competing cure reactions (Scheme II). Two of these reactions, the maleimide-maleimide cross-linking reaction (homopolymerization), reaction 4, and the Michael addition or amine-maleimide chain extension reaction (copolymerization), reaction 5, are widely believed to be the predominant reactions that occur in a thermal cure of these monomers [12,15-17]. The commonly held view is that reaction 5 predominates at low temperatures ($<180^\circ\text{C}$) and at higher temperatures ($180\text{-}220^\circ\text{C}$), reaction 4 occurs at a

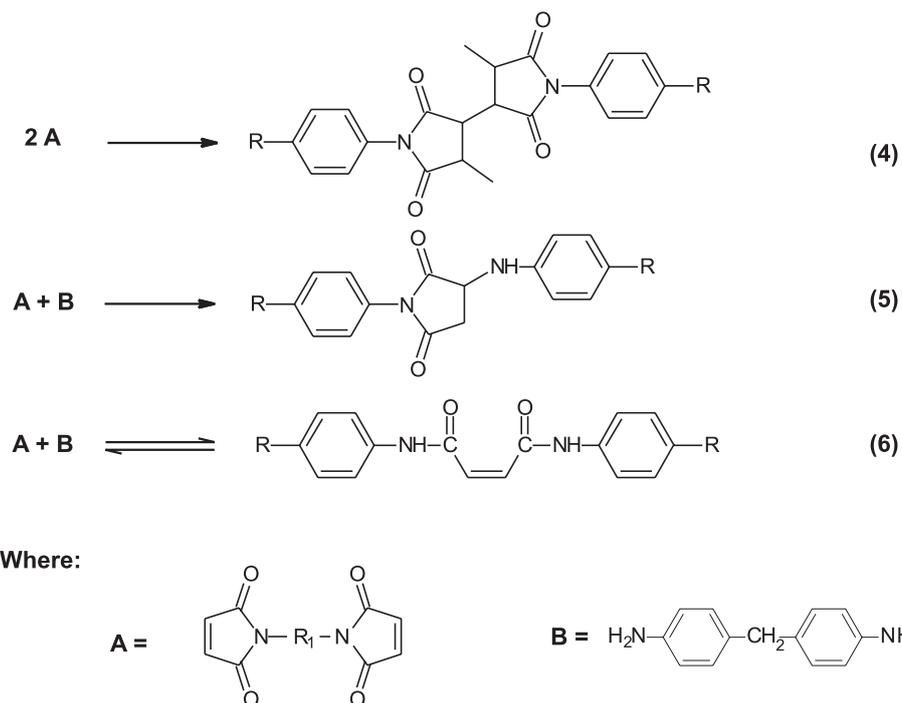
comparable rate [17]. The ring-opening aminolysis reaction, reaction 6, has been observed in certain solvents [16] and has been tentatively observed under normal cure conditions.

Thus, the dielectric properties are expected to change as a result of three effects, namely (1) the decrease in the permittivity owing to a decrease in the orientation polarization, when the dipole moments of the $-\text{NH}_2$ and epoxide groups are replaced by the dipole moments of the $-\text{OH}$ groups, (2) the decrease of the dipole moment per unit chain segment as the length of the chain and the density of cross-links increase, and (3) a rapid increase in the dipolar reorientation or dielectric relaxation time as the Brownian diffusion of the chain or network segments, carrying the dipoles becomes progressively slower or restricted, with the growth of the chain and the network [7].

Compare directly the DEA results of the samples, a single measurement frequency (1×10^4 Hz) which yielded well-defined loss peaks was chosen. As seen in the comparative plots of permittivity (ϵ') and loss factor (ϵ'') vs. temperature (Figures 1 and 2, respectively) for the four DGEBA/MDA/BMI systems (systems 1, 2, 3, and 4, respectively) the resin systems were heated from 20-200°C. The T_g of the uncured resin system 3 was detected at 40°C. At temperatures above 40-45°C (flow onset) the viscosity decreased as indicated by the increase in loss factor curves of all the four systems. In Figure 2, the loss factor curves reach a maximum at temperature values of 101.1, 98.99, and 96.45°C for the systems 2, 3, and 4, respectively, which are the points of the minimum viscosity, above which increasing molecular weight and network formation (due to the above mentioned number of reactions that may take place)



Scheme I. Usual epoxy:amine reactions.



Scheme II. Predominant reaction pathways for maleimide-amine systems: (4) maleimide homopolymerization reaction, (5) Michael addition reaction, and (6) ring-opening aminolysis reaction.

restricts ionic mobility and overshadows the temperature-induced fluidity. Thus, as a result, loss factor decreases. The cure reactions were completed at 140, 150, and 155°C for resin systems 2, 3, and 4, respectively, where permittivity values also became very low, because the dipoles could not move to align themselves with the electric field for the cross-linked resins. Similarly the ionic conductivity curves (Figure 3) reached their maximum values at temperatures 101.1, 99.94, and 96.14°C for the resin systems 2, 3, and 4, respectively. Above these temperatures the viscosity increased due to network formation. As a result, the movement of trace ions was hindered and decreases the conductivity data towards the completion of cure. The plots of permittivity, loss factor, and ionic conductivity vs. temperature for the four DGEBA/DDS/BMI systems, marked as 5-8 are given in Figures 4, 5, and 6, respectively. In Figure 4, double exotherm curing peaks, being more pronounced for systems 7 and 8 were observed. These resin systems were heated from 20-300°C. The T_g of the uncured system 6 was detected at 35.42°C (Figure 4). As temperature increased above 75°C, permittivity increased, reaching

maximum values at temperatures 145.5, (141.6 and 191.4), and (147.5 and 192.1)°C for resin systems 6, 7, and 8, respectively. In Figure 5, also with the onset of flow at 75°C, the loss factor curves increased, passing through maximum values (the points of minimum viscosity) at temperatures (146.5 and 168.1), (149.9 and 179.9) and (152.3 and 179.9)°C and then decreased towards the completion of curing reactions for resin systems 6, 7 and 8, respectively. The ionic conductivity curves vs. temperature (Figure 6) shows a frequency-independent increase with the decrease in the viscosity at temperatures above 75°C towards a maximum at temperature values of (145.8 and 168.8), (151.5 and 179.1) and (151.5 and 179.9)°C, followed by a decrease towards the completion of cure at 220, 225 and 230°C, respectively for the resin systems 6, 7, and 8.

The capacitive nature of the samples (DGEBA/MDA/BMI) increased with the addition of BMI which may be due to the introducing of more dipoles to the systems. Addition of various concentration of BMI to the above mentioned resin systems did not change the peak exotherm very much. Also increasing the concentration of BMI (5-15 wt %),

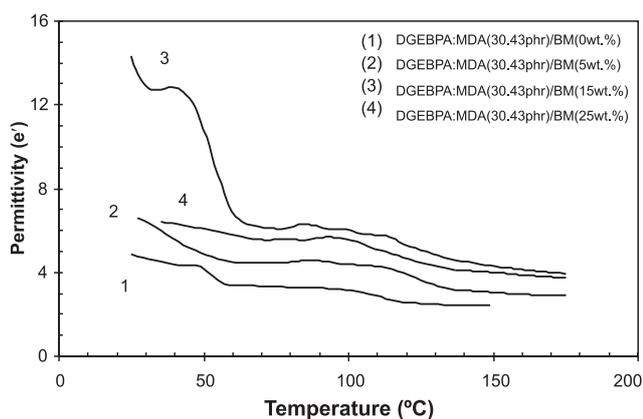


Figure 1. Comparative DEA heating profiles of permittivity of DGEBA/MDA/BMI systems at 1000 Hz.

increased the loss factor from 68.87 to 103.7, being much higher than that (27.41) of the resin system without any BMI content (Figure 2). This may be due to the presence of more free ions in the system by introducing BMI and increasing its concentration. However, the 25 wt % BMI content system showed a decrease in its loss factor (83.01) due to the excess of BMI. It results in taking place more reactions, thus more amount of viscosity exists during curing, which hindered the movement of free ions. The comparative ionic conductivity curves of these systems (Figure 3) also indicates that, the addition of BMI increased largely the ionic conductivity of the systems during curing (the reason being the same as explained for the loss factor data, before) where the conductivity values reaches to the maximum value of 38290, 57640

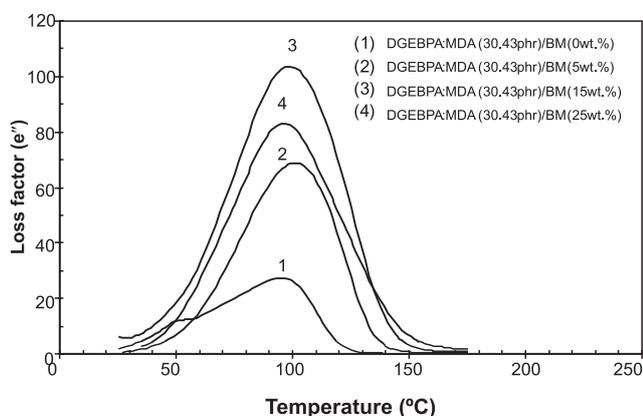


Figure 2. Comparative DEA heating profiles of loss factor of DGEBA/MDA/BMI systems at 1000 Hz.

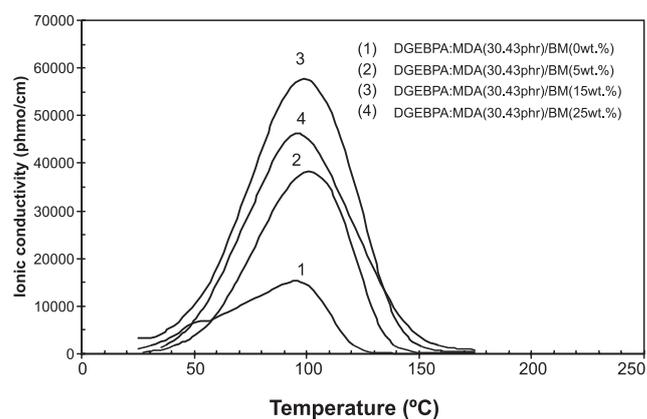


Figure 3. Comparative DEA heating profiles of ionic conductivity of DGEBA/MDA/BMI systems at 1000 Hz.

and 46160 pmho/cm for the systems with 5, 15 and 25 wt % BMI, respectively. In case of DGEBA/DDS/BMI systems permittivity decreases with the increase in the concentration of BMI (Figure 4) due to the number of reactions involved in the curing process as the concentration of BMI increased, resulting higher viscosity in the system and therefore hindering the movement of dipoles. In Figure 4, however permittivity improved in the case of 5 wt % BMI content system (43.23) as compared to that (36.51) of the system with no BMI content. It may be explained in terms of the presence of more dipoles in the system as the BMI was introduced. Figure 5 does not indicate any significant change in the peak exotherm temperature of these systems, however, a decrease in the loss factor values was

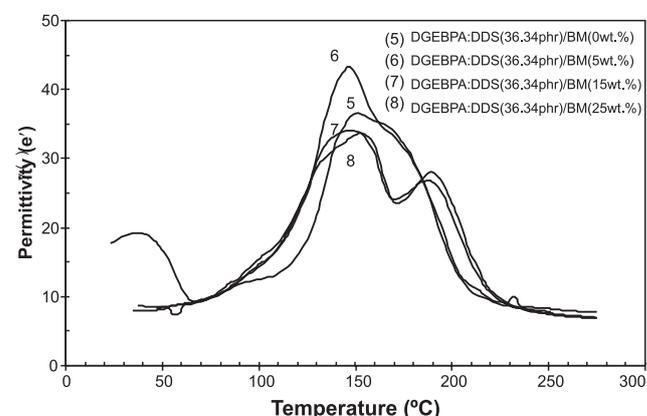


Figure 4. Comparative DEA heating profiles of permittivity of DGEBA/DDS/BMI systems at 1000 Hz.

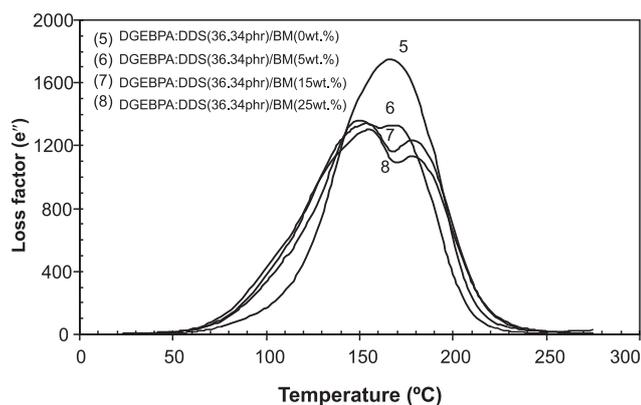


Figure 5. Comparative DEA heating profiles of loss factor of DGEBA:DDS/BMI systems at 1000 Hz.

observed with the addition of BMI to DGEBA/DDS system. In Figure 6, the ionic conductivity values observed at peak exotherm are 756400, 745000 and 723700 pmho/cm for the systems with 5, 15 and 25 wt % of BMI, respectively. The decrease in ionic conductivity with increase in the concentration of BMI, and also with respect to that (972700 pmho/cm) of the resin system without any BMI content, may be due to the more number of reactions taking place. It leads to an increase in the viscosity of the systems with more BMI content, thus, hindering the movement of free ions in the curing process.

The comparative plot (Figure 7) of temperature profiles of tan δ of the DGEBA/MDA(30.43 phr)/BMI with 0, 5, 15 and 25 wt % BMI laminates shows the

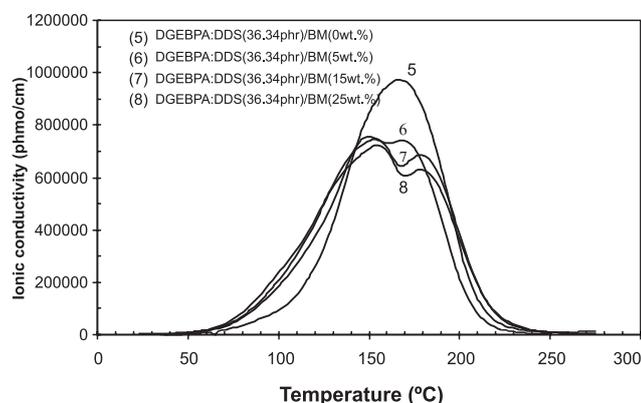


Figure 6. Comparative DEA heating profiles of ionic conductivity of GEBPA:DDS/BMI systems at 1000 Hz.

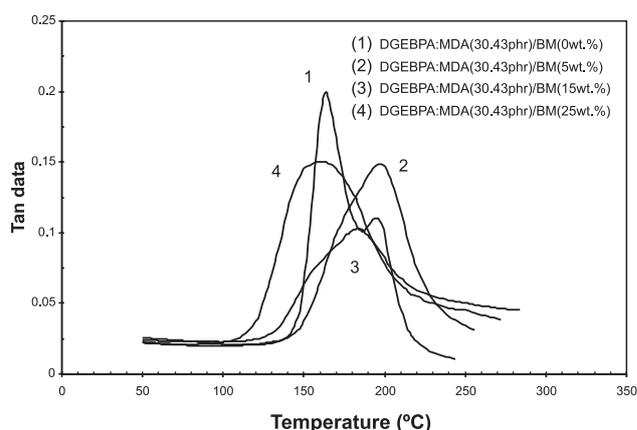


Figure 7. Comparative DMA temperature profiles of tan delta of glass fabric-reinforced DGEBA:MDA/BMI laminates.

characteristic major maximum tan δ characteristic of the α -transition which occurs at, or slightly above T_g . It is observed that, the addition and increasing of BMI content in the formulations from 5 to 15 and finally to 25 wt %, broadens the α -transition (T_g) peaks, which may indicate that, some inhomogeneity exists on a very small scale [18]. The tan δ curves also indicate that, the T_g of the laminates are increased by as much as 32 - 20°C, and suppressed in amplitude, by addition of 5 and 15 wt % BMI, respectively to the DGEBA/MDA(30.43 phr) system. It suggests that there is an increase in molecular weight and/or cross-linking. The $T_{g\infty}$ of the BMI containing systems are in the range 197-220°C, whereas that of the system without any BMI content is at 172°C. The flexural

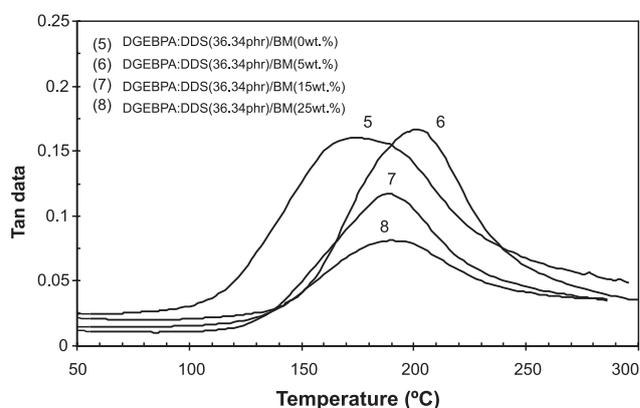


Figure 8. Comparative DMA temperature profiles of tan delta of glass fabric-reinforced DGEBA:DDS(36.34phr) /BMI laminates.

storage modulus (E') and shear storage modulus (G') data presented in Table 2 show an increase in the presence of BMI, and increasing the concentration of BMI from 5 to 15 wt % further increases the both storage moduli E' and G' . At lower temperatures, the resin system containing 25 wt % BMI shows almost the same values of flexural and shear storage moduli as the system with 5 wt % BM, but at elevated temperatures the moduli of the samples containing 25 wt % BMI drop below those of the system containing 5 wt % BMI.

Figure 8 is the plot of temperature profiles of $\tan \delta$ for DGEBA/DDS(36.34 phr)/BMI systems containing 0, 5, 15, and 25 wt % BM. It is observed that, T_g increases for the BMI containing resin systems, from a value of 172°C for the system without any BMI to 202°C for the 5 wt % BMI sample. Further, increasing the BMI concentration to 15 and 25 wt %, shows T_g values of 188 and 190°C, respectively with suppressed amplitudes as compared to the T_g 's of the other two systems. The broad α -transition (T_g) peaks specially for the systems with 0 and 25 wt % BMI indicate the existence of some inhomogeneity in these resin systems. $T_{g\infty}$ obtained, are in the range of 234-238°C for the BMI containing samples. That is about 20°C higher than that of the resin system without any BMI content.

CONCLUSIONS

The peak temperature of T_g of the uncured resin systems is dependent on the applied measurement frequency, which demonstrates this transition (T_g) is a relaxational or time-dependent event. It is due to the effects of the molecular dipoles, and as the measurement frequency increased (1×10^4 , 3×10^4 , 1×10^5 , 3×10^5 , and 1×10^6 Hz) the peak temperature increases. Thus, at temperatures near or below the T_g , dielectric properties of the resins are dominated by the molecular dipoles (frequency-dependent response) and at temperatures above T_g , The conductivity data curves merge together (frequency-independent response) and reach their maximum values of conductivity at curing peak temperature after which the viscosity increases due to network formation. As a result, the other movements of trace ions are hindered and decrease the conductivities towards the completion of cure.

Substitution of DDS for DMA as curing agent for DGEBA, increased dielectric properties during the curing process, and also increased the curing peak temperature. Addition of BMI to DGEBA/MDA systems, largely increased the ionic conductivity of the system during curing. However, the addition of BMI to DGEBA/ DDS resin systems, decreased the

Table 2: DMA results of glass fabric-reinforced DGEBA/ MDA (30.43 phr)/BMI and DGEBA/ DDS(36.34 phr)/BMI composites.

Resin system	Conc. of BMI resin (wt %)	Resin content (%)	T_g (°C)	$T_{g\infty}$ (°C)	Flexural storage modulus, E' (Gpa)				Shear storage modulus, G' (GPa)			
					75 (°C)	100 (°C)	125 (°C)	150 (°C)	75 (°C)	100 (°C)	125 (°C)	150 (°C)
DGEBA:MDA(30.43 phr)/BMI	0	25	164	172	14.6	14.1	13.6	12.1	5.1	4.9	4.7	4.2
	5	20	196	197	17.7	17.2	16.6	15.7	6.2	6.0	5.8	5.4
	15	20	184	220	22.4	21.6	20.7	18.0	7.8	7.5	7.2	6.3
	25	25	163	219	17.9	17.4	15.6	10.2	6.2	6.0	5.5	3.4
DGEBA:DDS(36.34 phr)/BMI	0	25	172	216	20.8	20.1	18.1	13.7	7.2	7.0	6.3	4.7
	5	25	202	234	20.7	20.1	19.5	18.2	7.2	7.0	6.8	6.3
	15	20	188	238	14.5	14.0	13.9	12.4	5.0	4.9	4.8	4.3
	25	25	190	236	22.0	21.2	20.3	18.5	7.6	7.4	7.1	6.4

ionic conductivity during cure. Double exothermic curing peaks were observed for DGEBA/DDS/BMI resin systems, become more pronounced with increasing the concentration of BMI.

Addition of bismaleimide (BM) resin in three different concentration to the glass-fabric DGEBA: MDA and DGEBA/DDS laminates, increased T_g , $T_{g\infty}$ and flexural and shear storage moduli (E' and G') considerably.

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