

# Rheological Study of Crosslinking and Gelation in Bismaleimide/Cyanate Ester Interpenetrating Polymer Network

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**ABSTRACT:** In searching for high performance polymer resins that have a combination of low dielectric constant and loss, high temperature resistance, ease of being processed, and other desirable properties, an interpenetrating polymer network (IPN) based on cyanate ester (CE) and 2,2'-diallylbisphenol A (DBA) modified bismaleimide resin (BMI) was prepared via prepolymerization followed by thermal curing. This work discusses the use of multiple waveform rheological technique to investigate the crosslinking and gelation behavior of this resin system at various temperatures. The gel point can be accurately determined from a single experiment using this technique. At the point of gelation, both the storage modulus ( $G'$ ) and loss modulus ( $G''$ ) of the IPN follow a similar power law equation with oscillation frequency used in the rheological measurement. Both the relaxation exponent  $n$ , a viscoelastic parameter related to the cluster size of the gel, and gel strength  $S$ , related to the mobility of the crosslinked chain segments, were determined via a curve fitting method. Both  $n$  and  $S$  were found to be temperature dependent in this BMI/DBA-CE IPN system. The apparent activation energy of gelation or curing reaction was found to be approximately 47.6 kJ/mol. © 2001 John Wiley & Sons, Inc. *J Appl Polym Sci* 80: 2437–2445, 2001

**Key words:** bismaleimide; cyanate ester; interpenetrating polymer network; gelation; multiwave rheological test; viscoelastic behavior of gel

## INTRODUCTION

Bismaleimide resins (BMIs) are good candidates for high performance resin matrix due to their good thermal stability, good fire resistance, low water absorption, and good retention of mechanical properties at elevated temperatures, especially in hot/wet environments. However, unmod-

ified BMI resins suffer from brittleness and poor processibility because of their high crosslink densities after curing, poor solubility in ordinary solvents, and high crystalline melting temperature of the monomers.<sup>1–4</sup> In addition, their relative high dielectric constants are not suitable for applications in electronic industries. Cyanate esters (CEs), with inherently low dielectric constant and loss dissipation factor, were postulated as potential modifiers for BMIs, but higher temperature (>250°C) is usually necessary in order to achieve crosslinking via cyclotrimerization of the cyanate ester groups in uncatalysed systems.<sup>5–7</sup> Previous studies showed that 2,2'-diallylbisphenol A

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(DBA) was one of most effective modifiers of BMI to improve the processibility and toughness. It can copolymerized with BMI through “Ene” and “Diels–Alder” reaction.<sup>8,9</sup> Another point is that the phenol groups in this allyl compound may act as a catalyst to the cyclotrimerization of cyanate esters.<sup>10</sup> Therefore, it is of interest to study resin systems based on blends of BMI/DBA and CE resins. In such systems, BMI/DBA and CE are crosslinked through different reactions to form two discrete networks which interpenetrated each other in the cured resin. This characteristic structure may result in synergistic effects on properties of the polymer networks, which are expected to couple the good dielectric properties of cyanate ester resin with the high temperature resistance and good mechanical and processing behavior of the modified BMI resin.

Gelation is one of the important properties of a thermoset resin. It occurs at a particular point during cure when an infinite and insoluble molecular network is formed.<sup>11,12</sup> Upon gelation, macroscopic flow becomes impossible and stress relaxation is difficult. Accurate determination of the gel point also would allow the estimation of the optimal temperature and time for which the sample should be heated before being allowed to set in the mold. Thus, both from scientific and technological viewpoints, it is necessary to be able to determine the gel point accurately and to understand the behavior of the resin upon gelation. Although extensive studies were carried out on gelation of a range of resins such as epoxy resin,<sup>13–15</sup> polyester,<sup>16,17</sup> and polyurethanes.<sup>18</sup> Little work has been report on resin system such as BMI/DBA–CE IPN. Hence it is useful to have a better understanding about gelation behavior of this new resin system. The present investigation encompassed the study of gelation behavior using rheology techniques. The gel point can be determined much more accurately by a multiwave test. The gel times at various temperatures can be used to calculate the activation energy of crosslinking. The obtained results will serve as a guideline to optimize the procedure of prepolymerization and selection of subsequent processing conditions.

### Rheology Theory

Rheological properties such as the viscosity and the dynamic moduli can be directly correlated to the evolving physical and mechanical properties of resin system during cure. A crosslinking poly-

mer at its gel point is in a transition state between liquid and solid. The gel point is defined unambiguously as the instant at which the weight average molecular weight diverges to infinity. In addition, the molecular weight distribution is infinitely broad ( $\overline{M}_w/\overline{M}_n \rightarrow \infty$ ), with molecules ranging from the smallest unreacted chains to the infinite, sample-spanning network clusters. The polymer reaches its gel point at a critical extent of crosslinking,  $\alpha \rightarrow \alpha_{\text{gel}}$ .<sup>19,20</sup>

Various rheological methods have been used to determine the gel point and they usually are based on definitions and approaches,<sup>6,21,22</sup> including the determination of the divergence point of the steady-state shear viscosity ( $\eta$ ) or normal stress ( $N_1$ ), the crossover point of the dynamic storage modulus ( $G'$ ) and loss modulus ( $G''$ ), the point at which the loss tangent ( $\tan \delta$ ) becomes independent of testing frequency ( $\omega$ ), or the power law relationship of the time-dependent relaxation modulus. However, some of these methods have some limitations and were often used incorrectly. For example, the crossover point of  $G'$  and  $G''$  coincides with gelation point only in certain special cases.<sup>11</sup>

It was proposed that stress relaxation behavior of a network polymer at the gel point follows a power law in previous studies<sup>23</sup>:

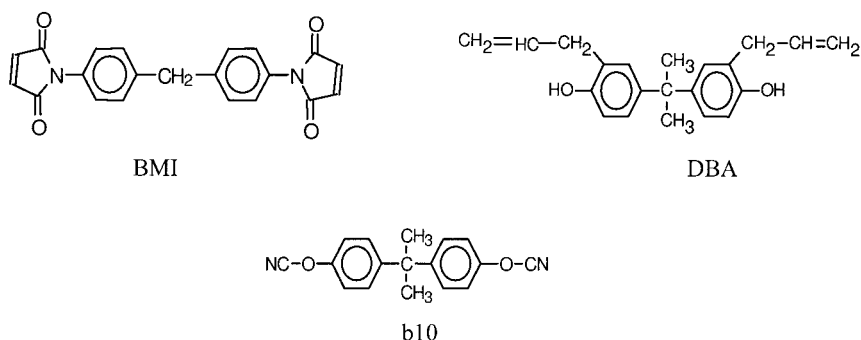
$$G(t) = St^{-n} \quad (1)$$

where  $S$ , the strength of the gel, depends on the flexibility of molecular chains and crosslinks and also on the crosslinking density at gel point. The relaxation exponent  $n$  ( $0 < n < 1$ ) is related to the geometry of clusters existing at the gel point. From the above equation, the frequency dependence of the dynamic shear moduli at gel point was deduced to be

$$G'(\omega) = \Gamma(1 - n)\cos(n\pi/2)S\omega^n \quad (2)$$

$$G''(\omega) = \Gamma(1 - n)\sin(n\pi/2)S\omega^n \quad (3)$$

Thus, the crossover of storage and loss moduli coincides with the gel point only for the resin system having a very specific exponent value of  $n$ . According to this definition, a resin reaches its gel point when  $G'$  is equal to  $G''$ , and this is only possible when  $n$  is 0.5 and  $\tan \delta$  is equal to 1. However, the exponent  $n$  of many thermosetting resins may deviate substantially from the value of



**Scheme 1** Structures of raw materials.

0.5. Therefore, this definition is often invalid and a more general approach has to be adapted.

In any case, eqs. (2)–(3) are generally valid for linear viscoelastic polymer gels and the loss tangent  $\tan \delta$  should be frequency independent at the gel point because

$$\tan \delta = G''/G' = \tan(n\pi/2) \quad (4)$$

If the evolution of  $\tan \delta$  with cure time was measured at different frequencies, the various curves would coincide at a single point, corresponding to the gel point. This can be obtained via the use of multiwave rheology tests.<sup>11,15</sup>

$$\gamma = \sum_{i=1}^m \gamma_i \sin(\omega_i t) \quad (5)$$

Where  $\omega_1 = \omega_f$ ,  $\omega_2 = m_2 \omega_f$ ,  $\omega_3 = m_3 \omega_f$ , ...  $\omega_i = m_i \omega_f$  and  $m_2, m_3, \dots, m_i$  are integers.

The strain applied  $\gamma$  is the sum of the Fourier series described by each individual strain at different frequency. The frequencies chosen are harmonics (integer multiples) of a fundamental frequency  $\omega_f$ . Equation (6) expresses the condition requiring that the sum of the individual strain amplitudes has to be below the critical amplitude  $\gamma_c$ , corresponding to the linear viscoelastic limit.

$$\sum_{i=1}^m \gamma_i \leq \gamma_c \quad (6)$$

The response from this multiwave oscillation is deconvolved by Fourier transformations into separate responses due to the individual frequencies. From the results of the multiwave experiment, the gel point can be measured by detecting the

point at which the loss tangent becomes frequency independent.

## EXPERIMENTAL

### Prepolymer Preparation

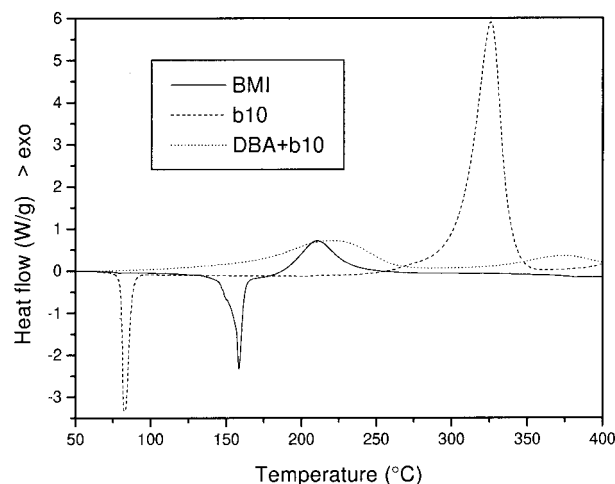
BMI (95%) and DBA (85%) were obtained from Aldrich Chemicals and were used without further purification. The 2,2'-bis(4-cyanatophenyl) isopropylidene (b10, 99%) with melting point 79°C was supplied by Ciba-Geigy of the United States and was used as received. (See Scheme 1.) BMI and DBA with molar ratio (2 : 1) were mixed with stirring at 120 ~ 150°C for 25 min to form a homogeneous mixture. Appropriate cyanate ester b10 (BMI/DBA: b10 = 75 : 25, weight ratio) was added into the above mixture at 120°C and the final mixture was kept at 140°C for about 10 min before degassing. The prepolymer (coded bb72) obtained was transparent and was stored in a freezer until being used.

### Differential Scanning Calorimetry Analysis of Monomers and Prepolymer

Differential scanning calorimetry (DSC) was performed at a heating rate of 10°C/min from room temperature to 400°C using a TA instruments Model 2010 calorimeter. Weighed monomers or prepolymer (around 5 mg) were analyzed in hermetically sealed aluminum pans under a nitrogen purge gas.

### Rheological Measurements

A Rheometric Scientific ARES rheometer was used to determine the gel point of the BMI/DBA-CE IPN resin system. The rheometer was



**Figure 1** Overlay DSC plots of BMI, cyanate ester (b10), and mixture of DBA + b10.

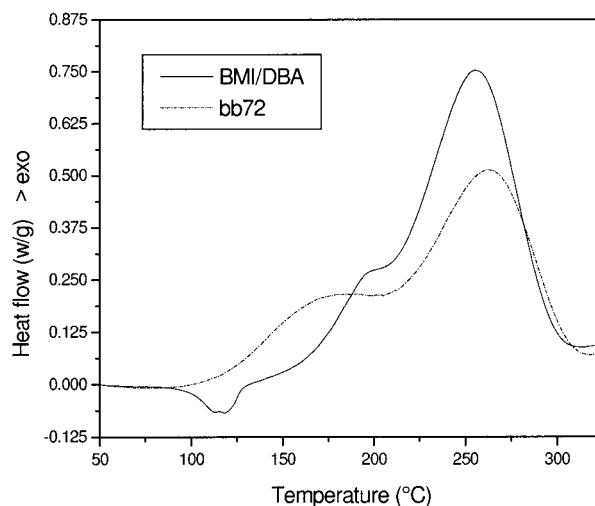
used in parallel-plate configuration [diameter ( $D$ ) = 40 mm, gap ( $H$ ) = 0.5 mm] at temperatures of 130, 140, and 150°C. Superimposed signals of 0.2, 1.0, 5.0, 10.0, and 20.0 Hz with a strain ( $\gamma$ ) of 2% were used at each frequency in multiwave test. Data for  $G'$  and  $G''$  as a function of curing time can be plotted simultaneously from the multiwave dynamic rheological tests. For steady-state shear tests, low rotation speed of 0.1 rad s<sup>-1</sup> was used and the steady-state shear viscosity  $\eta$  or the normal stress  $N_1$  were recorded as function of cure time. All tests were conducted in air. The temperature of the sample was monitored by a K-type thermocouple that was inserted through the center of the bottom shaft, and its tip was 1 mm from the shear surface of the bottom plate. The temperature reached the set point within 1 min after sample insertion, through adjustable temperature control parameters following a warm-up time of 15 min prior to the sample placement in the rheometer. The steady state temperature was kept to within 0.5°C of the set point.

## RESULTS AND DISCUSSION

### DSC Analysis

Figure 1 shows the DSC thermograms of the two monomers (pure BMI and cyanate ester b10) and the mixture of b10 and DBA. The crystalline melting endotherm of pure BMI occurs at 158°C, while the homopolymerization spans between 180 ~ 300°C, reaching an exothermic peak at 214°C. The curing onset and exothermic peak tempera-

ture of pure b10 homopolymerization occur at 306 and 327°C, respectively. The high curing temperature is undesirable from the processing point of view. An earlier report<sup>24</sup> showed that phenol groups have catalysis effect to cyanate ester cyclotrimerization. In order to distinguish the catalysis effect of OH groups in DBA, thermal behavior of the mixture DBA + b10 were characterized by dynamic DSC from room temperature to 400°C at a heating rate of 10°C/min, results shown in Figure 1. It can be seen that the onset and peak temperatures for curing reactions of b10 reduce significantly by about 100°C. This effect was also confirmed by the reaction heat of b10 curing in the mixture DBA + b10. The reaction heat ( $\Delta H$ ) of pure b10 self-crosslinking was estimated as 764 J/g. The enthalpy ( $\Delta H$ ) value of the first peak which corresponds to b10 self-crosslinking in mixture DBA + b10 (b10 by weight 52%, same OH groups concentration with bb72) were calculated as 402.1 J/g, as against the theoretical value 406.2 J/g, which shows a close match. Therefore, the DSC results have provided strong evidence that DBA is an effect catalyst for b10 curing in this resin system. Figure 2 shows that, in the BMI/DBA-CE blend system (prepolymer bb72), the onset curing temperature is substantially reduced and there is no significant exotherm observed above 300°C. It was found that the glass transition temperature ( $T_g$ ) of the cured resin system could not be detected effectively using the DSC method probably due to their high degree of crosslinking. However, the  $T_g$  can be measured using DMA technique. The glass transition temperatures of cured BMI/DBA, b10 and their blend



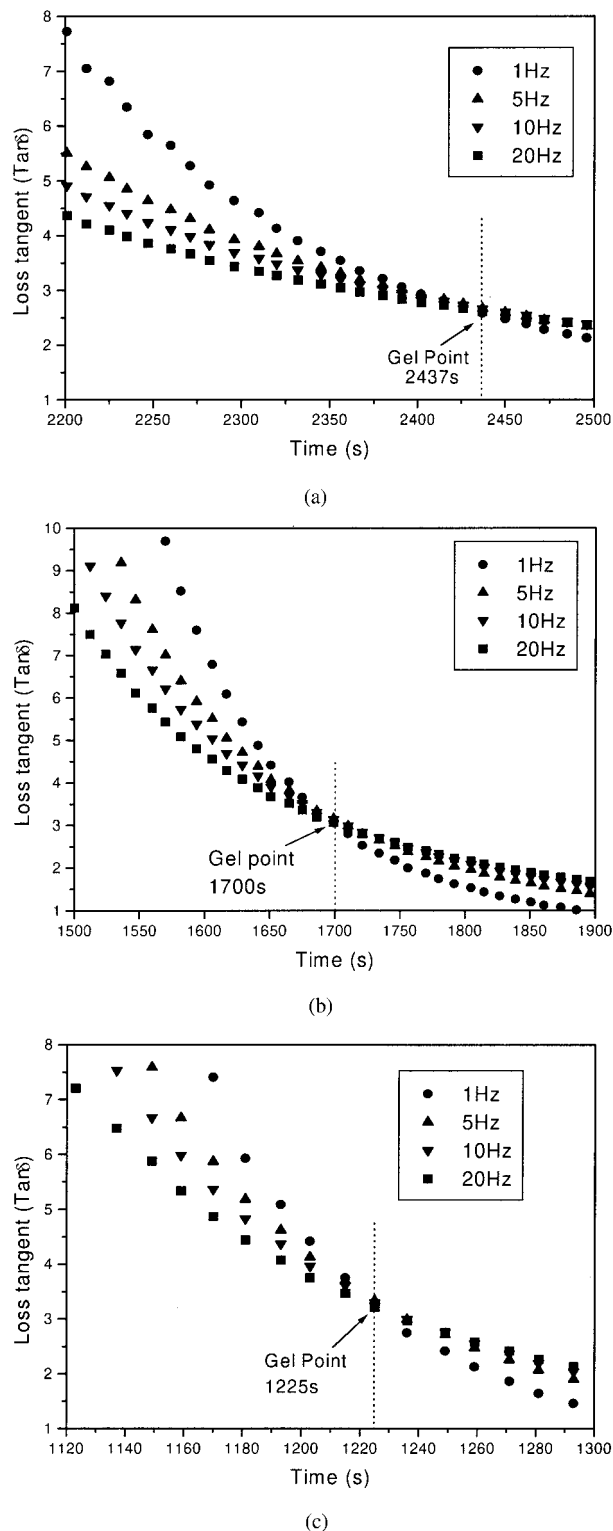
**Figure 2** Overlay DSC plots of BMI/DBA and bb72.

system bb72 are 335, 308, and 282°C, respectively. The lower  $T_g$  of blend system may attribute to the less crosslinking density resulted by interlock effect of IPN resin system.

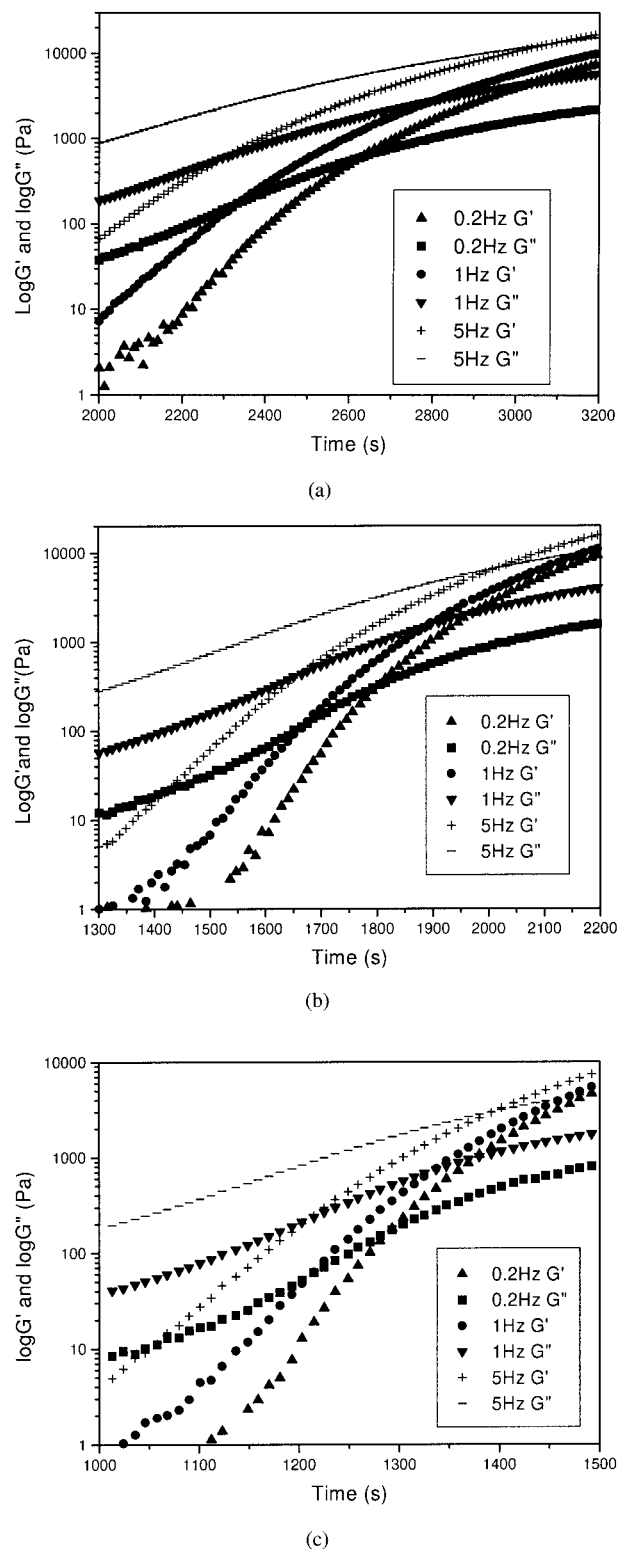
### Rheological Study

A comparison of the gel point of this BMI/DBA-CE IPN resin system, measured by a multiwave test and a steady-state shear test is shown in Figures 3–5 and Table I. Figure 3 presents the results of a multiwave test for the BMI/DBA-CE IPN resin system, showing the  $\tan \delta$  vs cure time plots at 1, 5, 10, and 20 Hz at 130, 140, and 150°C. In this test, the loss tangent is initially undetectable due to the small torque values below the resolution of the transducer. Near the gel point, however, the torque becomes measurable, and the gel point can be determined precisely from the position at which  $\tan \delta$  becomes independent of frequency [2437, 1700, and 1225 s; refer to Figs. 3(a), (b), and (c)]. Although the  $G'$  and  $G''$  crossover point was often used to define the gel point of resin system, in this case such definition is clearly invalid for the BMI/DBA-CE IPN resin system. Figure 4 shows the evolution of the storage ( $G'$ ) and the loss ( $G''$ ) moduli of BMI/DBA-CE IPN resin system during curing process at the isothermal temperature of 130, 140, and 150°C. It can be seen that the crossover point of  $G'$  and  $G''$  of this resin system shows strong frequency dependence at each selected temperature. The crossover point of  $G'$  and  $G''$  appeared later with increasing frequency. Detailed comparison between Figure 3 and Figure 4 reveals that at the same curing temperature, the  $G'$  and  $G''$  crossover point always occurs later than the gel point determined by the point at which  $\tan \delta$  becomes independent of frequency. As discussed previously,<sup>11</sup> the gel point can only be determined accurately at some special situation using crossover point of  $G'$  and  $G''$ . This condition is that the stress relaxation in the critical gel follows a power law, and the exponent  $n$  of which must be exactly 0.5. For most network systems at gel point, though stress relaxation also follows a power law, the relaxation exponent has a different value, i.e.,  $n \neq \frac{1}{2}$ . Gel point clearly does not coincide with the  $G'$ - $G''$  crossover in such cases. It was reported that the gel point occurs earlier than the  $G'$  and  $G''$  crossover for epoxy resins<sup>20,25</sup> with  $n$  values higher than 0.5. This is consistent with the results for this BMI/DBA-CE resin system.

Figure 5 plots the  $\log G'$  and  $\log G''$  against  $\log \omega$  at gel point for different curing temperatures at

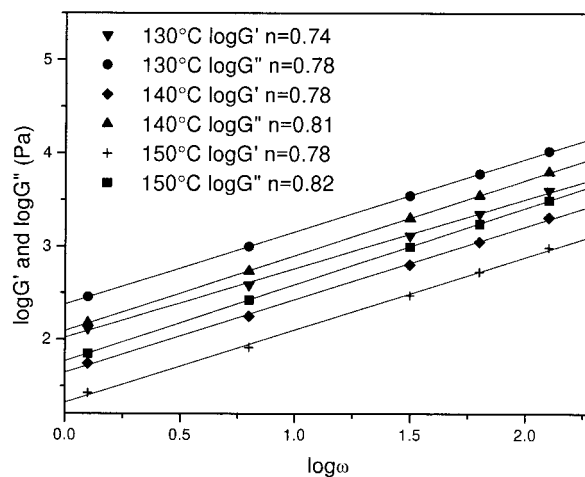


**Figure 3** Loss tangent as a function of cure time for BMI/DBA-CE IPN resin system at  $f = 1, 5, 10,$  and  $20$  Hz,  $\gamma = 2\%$ , and (a)  $T = 130^\circ\text{C}$ , (b)  $T = 140^\circ\text{C}$ , (c)  $T = 150^\circ\text{C}$ .



**Figure 4** Evolution of the storage modulus ( $G'$ ) and loss modulus ( $G''$ ) for BMI/DBA-CE IPN resin system at  $f = 0.2, 1,$  and  $5$  Hz,  $\gamma = 0.2\%$ , and (a)  $T = 130^\circ\text{C}$ , (b)  $T = 140^\circ\text{C}$ , (c)  $T = 150^\circ\text{C}$ .

130, 140, and  $150^\circ\text{C}$ . The relaxation exponent  $n$  can be obtained from the slope of plotting  $\log G'$  and  $\log G''$  vs  $\log \omega$ . They are shown in the graph and also in Table II. It can be seen that the  $n$  values obtained from  $\log G' - \log \omega$  plots are slightly but consistently higher than those from  $\log G'' - \log \omega$  plots at all tested temperatures, although they are expected to be identical according to eqs. (2) and (3). This difference can be attributed to the possible deviation from the linear viscoelastic behavior assumed in the theory. Also, there seems to be a small increasing trend in the  $n$  values with temperature, indicating increasing viscous component in the system at higher temperature. This is reasonable in a polymer system. Although further investigations are required in order to fully understand the phenomenon, this temperature dependence of relaxation exponent at gel point indicates that the cluster size in the gel is related to the curing temperature. It seems that curing at high temperature may have resulted in smaller cluster size and hence higher viscous component in the viscoelastic BMI/DBA-CE gel indicated by higher  $n$  value. According to eqs. (2) and (3), the gel strength  $S$  can be calculated from the interception of  $\log G'$  and  $\log G''$  vs  $\log \omega$  plots by showing a  $\gamma$  function. The  $S$  values are listed in Table II. Although no vigorous theoretical interpretation of  $S$  appears to exist, it is believed that gel strength reflects the mobility of chain segments at the gel point. With increasing temperature, gel strength  $S$  decreases due to enhanced chain mobility at higher temperature in this case. As mentioned earlier, different cluster sizes at different temperatures may also contribute to decreasing  $S$ . Discrepancies are also seen in the  $S$  values obtained



**Figure 5** The frequency spectrum of  $G'$  and  $G''$  at the gel point of 130, 140, and  $150^\circ\text{C}$ .

**Table I Gel Time Determined by Different Rheological Techniques for BMI/CE Resin**

Test	Conditions	130°C, Gel Time (s)	140°C, Gel Time (s)	150°C, Gel Time (s)
Multiwave test	$f = 0.2, 1, 5, 10, 20$ Hz	$2437 \pm 50$	$1700 \pm 50$	$1225 \pm 50$
Tan $\delta$ independent of frequency				
Crossover point of $G'$ and $G''$	0.2 Hz	$2630 \pm 50$	$1810 \pm 50$	$1290 \pm 50$
	1 Hz	$2805 \pm 50$	$1892 \pm 50$	$1342 \pm 50$
	5 Hz	$3090 \pm 50$	$2015 \pm 50$	$1398 \pm 50$
Steady time-infinite viscosity or normal stress	$\omega = 0.1$ rad s <sup>-1</sup>	$2200 \pm 200$	$1490 \pm 200$	$1180 \pm 200$

from  $\log G' - \log \omega$  and  $\log G'' - \log \omega$  plots. This is again attributed to the deviation from linear viscoelastic behavior of the gel.

Steady-state shear tests determine the gel time as the time where the viscosity difference becomes infinite. Although this method has some limitation in accuracy, it does have some advantages, such as its simplicity and the low strain applied resemble that of existing resin casting. The gel time can be estimated from plots of  $1/\eta$  vs time as shown in Figure 6. These gel points were found to be about 2200, 1490, and 1180 at 130, 140, and 150°C, respectively. Typically the shear viscosity rises as gelation commences, though in some cases where the heat of reaction is sufficient to raise the resin temperature, the viscosity value might actually drop initially. As the gelation process proceeds, the viscosity value would rise exponentially and provide a demarcation in plot of the viscosity against the cure time. Of interest in the steady-state shear test is that the first normal stress difference ( $N_1$ ), which can measure the elasticity of the resin system, also provides a good indication of the gel point. It was noted that a large decrease in the normal stress occurred during a steady-state shear test near the gel point. This may be due to the shrinkage and increased adhesion of the resin that drew the plates together as the material gelled. In resins where shrinkage is much less, it is more probable that the gel point would signify an increase in the normal stress due to the increased elasticity of the material. Good agreement of gel points of this resin system is obtained from both  $\eta$  and  $N_1$  measurements, which demonstrated that the increase in viscous drag during gelation is clearly paralleled with increase in elasticity of the growing network, and provides another useful tool for gel point determination.

### Activation Energy of Crosslinking

It has been shown in the literature that apparent activation energy for the crosslinking reaction can be calculated from the measurement of the gel time at different temperatures.<sup>15,26</sup> The assumption is typically made that the curing kinetics involves only a single reaction with single activation energy. While this is often not the case, the single activation energy value so obtained can be considered to be representative of the multiple reactions in progress. Since the network structure is a unique function of conversion, the conversion at the gel point may be considered constant and independent of temperature. The gel time–temperature profile follows an Arrhenius relationship. The kinetic equation can then be integrated in the following manner:

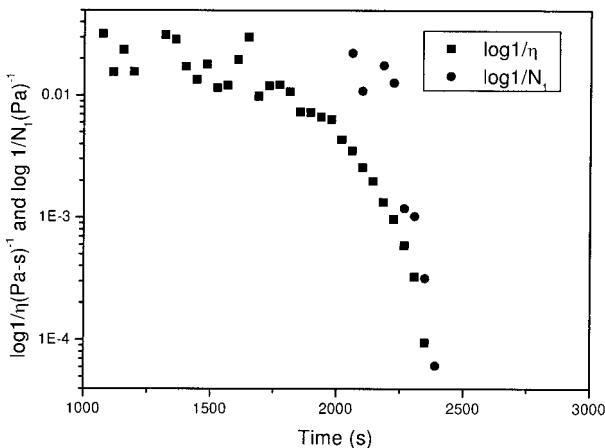
$$d\alpha/dt = kf(\alpha); k = Ae^{-E/RT}$$

$$\ln t_{\text{gel}} = \left\{ \ln \left[ \int_0^{\alpha_{\text{gel}}} \frac{d\alpha}{f(\alpha)} \right] - \ln A \right\} + \frac{E}{RT} \quad (7)$$

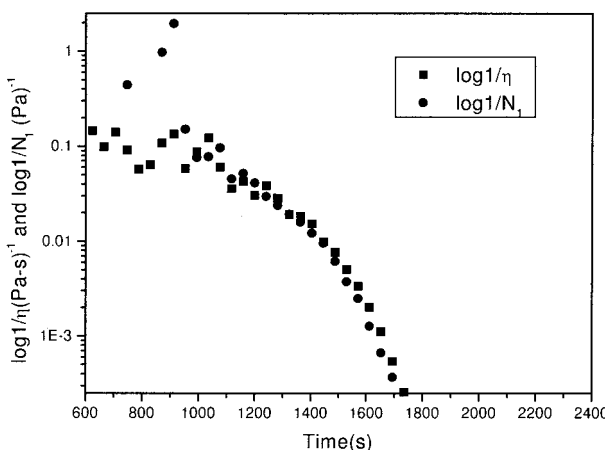
Table I gives the gel time determined by multiwave rheological test at 130, 140, and 150°C. As

**Table II The Obtained  $n$  and  $S$  Value at the Gel Point of Different Temperature**

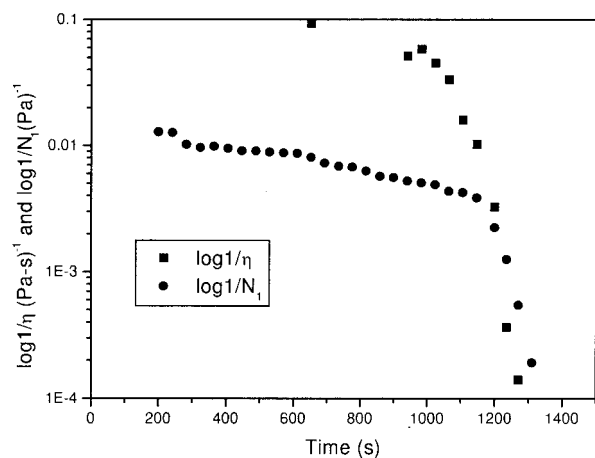
Temperature (°C)		$n$	$S$
130	$G'$	0.74	75.66
	$G''$	0.78	60.93
140	$G'$	0.78	31.38
	$G''$	0.81	26.65
150	$G'$	0.78	14.88
	$G''$	0.82	11.86



(a)



(b)



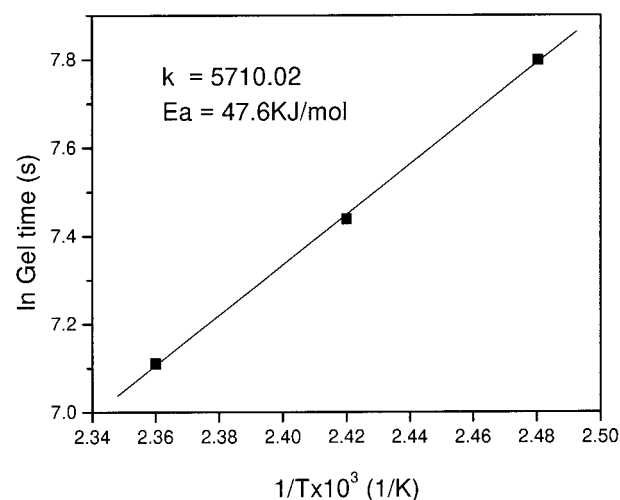
(c)

**Figure 6** Steady time test ( $\omega = 0.1 \text{ rad s}^{-1}$ ) for BMI/DBA-CE IPN resin system at (a)  $T = 130^\circ\text{C}$ , (b)  $T = 140^\circ\text{C}$ , and (c)  $T = 150^\circ\text{C}$  using parallel plates with a diameter of 40 mm and a gap of 0.5 mm.

expected the gel time decreased with increasing temperatures. The activation energy  $E$  of the curing can be calculated from the slope of a semilogarithmic plot of the gel time against  $(1/T)$ , as shown in Figure 7. The value for this BMI/DBA-CE IPN resin system is found to be approximately 47.6 kJ/mol. This result is comparable to the value (53.5 kJ/mol) obtained by a method using DSC (results not shown). After obtaining the value for the curing activation energy of this resin system, eq. (7) also can be used to estimate the gel time of any temperature.

## CONCLUSION

IPN based on BMI and CE have been prepared via prepolymerization followed by thermal curing. Multiwave rheological technique has been used to measure isothermal gel times in BMI/DBA-CE IPN resin systems. The multiwave rheological technique was found to be effective and can be used to generate frequency spectra during curing of this IPN resin. The gel points of this resin system have been accurately determined using the point at which  $\tan \delta$  becomes independent of frequency at different temperatures. The storage modulus  $G'$  and loss modulus  $G''$  followed the power law at the gel point; the scaling exponent  $n$  was found to increase with temperature indicating higher viscous component in the viscoelastic polymer gel at higher gelation temperature. The gel strength  $S$ , which reflects the mobility of chain segments, was found to decrease with increasing temperature probably due to smaller



**Figure 7** Semilogarithmic plot of gel time vs  $1/T$ .



clusters resulted from high temperature curing. The properties of the gel of the IPN system are of interest since they may be closely linked to those of the bulk properties of the cured resin. Also, the activation energy for the cure reaction of this resin system has been determined to be 47.6 kJ/mole using this multiwave method.

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## REFERENCES

1. Wang, C.; Hwang, H. *J Appl Polym Sci* 1996, 60, 857.
2. Dao, B.; Hawthorne, D. G.; Hodgkin, J. H.; Jackson, M. B.; Morton, T. C. *High Perform Polym* 1996, 8, 245.
3. Stenzenberger, H. D.; Romer, W.; Herzog, M.; Konig, P.; Fear, K. Proceeding of the 10th International European Chapter Conference of the SAMPE, Birmingham, United Kingdom, July 11–13, 1989.
4. Wang, C.; Hwang, H. *Polymer* 1996, 37, 499.
5. Barton, J. M.; Hamerton, I.; Jones, J. R. *Polym Int* 1992, 29, 145.
6. Barton, J. M.; Hamerton, I.; Jones, J. R. *Polym Int* 1993, 31, 95.
7. Jeffrey, T. G.; Bernd, K. A. *IBM J Res Develop* 1988, 32, 616.
8. Stenzenberger, H. D.; Konig, P.; Romer, W.; Herzog, M.; Breitigam, W. V. 36th International SAMPE Symposium, April 15–18, 1991.
9. Liang, G.; Gu, A.; Lan, L. *J Adv Materials* 1996, 27, 61.
10. Hamerton, I. *Chemistry and Technology of Cyanate Ester Resins*; Blackie Academic and Professional Press: London, 1994; Chap 3.
11. Winter, H. H. *Polym Eng Sci* 1987, 27, 1698.
12. Yeong-Show, Y.; Laurent, S. *Polym Eng Sci* 1991, 31, 321.
13. Martin, J. E.; Adolf, D.; Wilcoxon, J. P. *J P Phys Rev A* 1989, 39, 1325.
14. Lipshiz, S.; Macosko, C. W. *Polym Eng Sci* 1976, 16, 803.
15. Oyanguren, P. A.; Williams, R. J. *J Appl Polym Sci* 1993, 47, 1361.
16. Rubinstein, M.; Colby, R. H.; Gillmor, J. R. *Polym Prepr* 1989, 30, 81.
17. Yang, Y. S.; Suspene, L. *Polym Eng Sci* 1991, 31, 321.
18. Durand, D.; Delsanti, M.; Adam, M.; Luck, J. M. *Europhys Lett* 1987, 3, 297.
19. Holly, E. E.; Venkatraman, S. K.; Chambon, F.; Winter, H. H. *J Non-Newtonian Fluid Mech* 1988, 27, 17.
20. Hsu, S. H.; Jamieson, A. M. *Polymer* 1993, 34, 2602.
21. Matejka, L. *Polym Bull* 1991, 26, 109.
22. Yang, Y. S.; Suspene, L. *Polym Eng Sci* 1991, 31, 321.
23. Scanlan, J. C.; Winter, H. H. *Macromolecules* 1991, 24, 47.
24. Bauer, M.; Bauer, J.; Kühn, G. *Acta Polym* 1986, 37, 715.
25. Halley, P. J.; Mackay, M. E.; George, G. A. *High Perform Polym* 1994, 6, 405.
26. Gough, L. J.; Smith, I. T. *J Appl Polym Sci* 1960, 9, 362.