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Function and performance of silicone copolymer. Part IV. Curing behavior and characterization of epoxy–siloxane copolymers blended with diglycidyl ether of bisphenol-A

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Abstract

In this research, a siloxane-type epoxy resin (SG copolymer), which has pendant epoxide rings on the side chain of the polysiloxane polymer backbone, was synthesized by the hydrosilylation reaction of poly(methylhydrosiloxane) with allyl glycidyl ether. The polymer structures were characterized by ^{1}H NMR. The SG resin was then blended with a commercial epoxy resin (diglycidyl ether of bisphenol-A, DGEBA) at various ratios, using dicyandiamide (DICY) as a curing agent. The curing behaviors were studied by DSC. It was found that the initial curing temperature (T_i) and peak curing temperature (T_p) were increased by the addition of SG copolymer to the epoxy resin. Their morphology, mechanical properties and the stability of the cured piece were investigated using SEM, DMA and TGA, respectively. The results show that the addition of SG copolymer increases the mobility of the crosslinked network, and increases the thermal stability. © 2000 Elsevier Science Ltd. All rights reserved.

Keywords: Diglycidyl ether of bisphenol-A; Siloxane-type epoxy; Dicyandiamide

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1. Introduction

Epoxy resins, one of the most important classes of thermosetting polymers, are widely used for coatings, adhesives and advanced materials [1–3]. Because of their excellent resistance to water, heat and chemicals, and their excellent electrical and mechanical properties, epoxy resins are also widely used as molding compounds or encapsulation materials [4,5] for electronic components. However, highly crosslinked epoxy resins are relatively brittle; thus, a second component such as a rubber or a thermoplastic modifier is added to enhance the fracture toughness of the epoxy resin. Most of the earlier studies on the toughening of epoxy resins have been carried out by utilizing carboxy- or amine-terminated acrylonitrile–butadiene rubbers, functionally terminated acrylates, poly(phenylene oxide) and alkylene oxides [6–9]. Recently, the introduction of incompatible materials such as siloxanes to epoxy resins [10–14] is attracting more and more attention in this field because siloxanes possess many unique properties, such as low glass transition temperature, low surface tension, good flexibility, non-flammability and high resistance to thermal oxidation.

In rubber modification of epoxies using silicone resins, the phase separation of the siloxane component from the epoxy matrix triggers an extrinsic toughening mechanism that effectively shields the growing crack tip, thus improving the fracture toughness [15–17]. However, compounding the two incompatible silicon- and carbon-based ingredients in the epoxy formulations always cause problems. Therefore, it is interesting to synthesize a polymer that possesses both the polysiloxane and epoxide moieties.

In this study, a siloxane-type epoxy (SG copolymer) which is a polysiloxane copolymer containing a epoxide group on the side chain was synthesized from the poly(methylhydrosiloxane), and the resulting SG copolymer was mixed with an epoxy resin (DGEBA) to modify the characteristics of the latter. Since the epoxide group appears in the structure of SG molecule, the compatibility between SG copolymer (silicone-based) and DGEBA epoxy (carbon-based) is very good and they form a clear solution when they are blended together at room temperature. In the experiment, the difference in the reactivity between SG copolymer and DGEBA epoxy resin and the morphology of cured SG/DGEBA blends prepared at various ratios were studied by DSC, DMA and SEM. Also, the resistance of the cured

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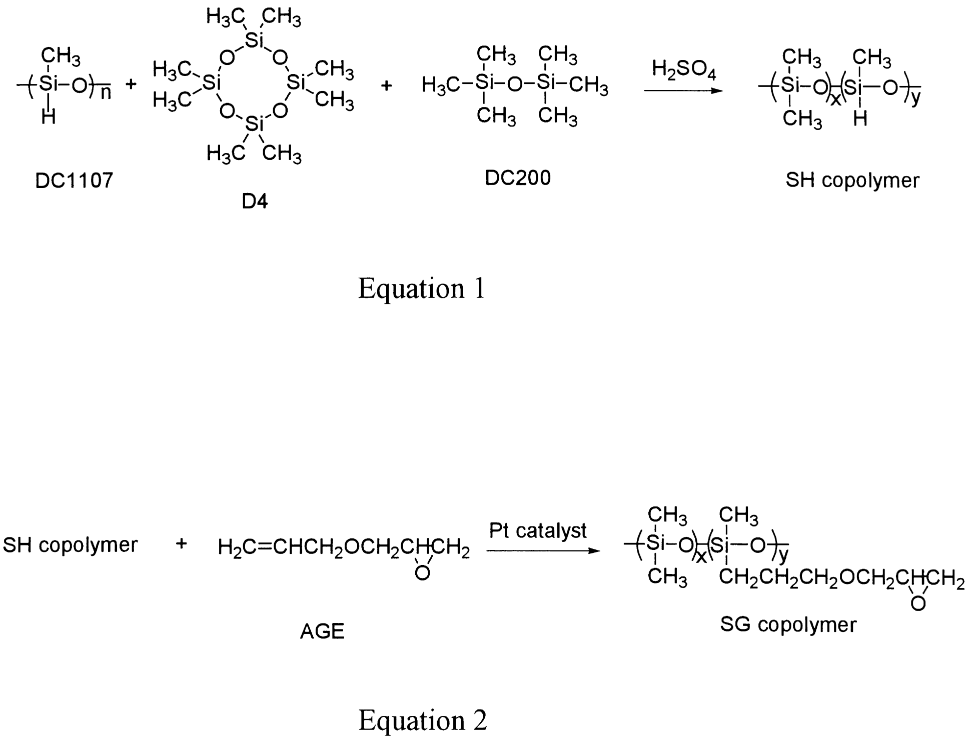


Equation 1

Equation 2

Scheme 1.

blends to thermal degradation was investigated by TGA and was found to be improved by the addition of SG copolymer to the epoxy matrix.

2. Experimental

2.1. Materials

Octamethylcyclotetrasiloxane (D4), polymethylhydrogensiloxane (DC1107), hexamethylcyclotetrasiloxane (DC200), and platinum divinyltetramethyl disiloxane complex (Pt-catalyst) were purchased from Dow Corning Corporation; diglycidyl ether of bisphenol-A (DGEBA, Nan-Ya), allyl glycidyl ether (AGE, Acros), dicyandiamide (DICY, TCI), *N*,*N*-benzyldimethylamine (BDMA, Acros), sulfuric acid (UCW Ltd.), toluene (Alps), 1,4-dioxane (Alps), methanol (Alps), NaOH (UCW Ltd), ethylene glycol monoethyl ether (Methyl Cellosolve, MC) (Tedia), and sodium hydrogen carbonate (Tedia) were used as received. All solvents were reagent grade or were purified by standard methods.

2.2. Synthesis of siloxane-type epoxy resin (SG copolymer)

To a 500 ml four-necked reactor, DC1107 (80 g), D4 (17.5 g) and DC200 (2.5 g) were added. After raising the temperature to 45°C, sulfuric acid (5 g) was added and stirred under nitrogen atmosphere for 48 h. Sodium hydrogen carbonate was then added to neutralize the acid in the reaction mixture. The precipitate was filtrated out with filter paper, and the product was charged into a 500 ml flask with an excess of AGE (1.05 equivalents of Si–H), and 100 g of toluene. After raising the temperature to 75°C, 1 wt% of Pt-catalyst was added to the flask along with a low stream of nitrogen and stirred until the absorption peak of the Si–H peak (ca. $2165\ \mathrm{cm}^{-1}$) in the infrared spectra disappeared. The reaction mixture was then placed in a rotary evaporator to remove excess AGE and the solvent. The structure of SG-copolymer was confirmed by ^{1}H NMR.

2.3. Epoxy equivalent weight determination

A 0.2 N HCl/dioxane solution was prepared by placing 10 ml of methanol and 3.2 ml of HCl (35%) in a 200 ml volumetric flask, and then adding 1,4-dioxane to a level of 200 ml. NaOH (2 g) was dissolved in 50 ml of water in a 500 ml flask; then a mixture of methyl cellosolve (MC, 150 ml) and methanol (350 ml) were added into the flask to obtain a 0.1 N NaOH solution. Next, EEW titration was performed by dissolving SG copolymer (*W* g) in 20 ml dioxane. HCl/dioxane solution (0.2 N; 27 ml) was then added and mixed well. The mixture was heated to 40°C for 2 h, and then cooled to room temperature. The obtained solution was titrated with 0.1 N NaOH solution using cresol red as the indicator. The same steps were executed with the blank solution, and the EEW was calculated using the following

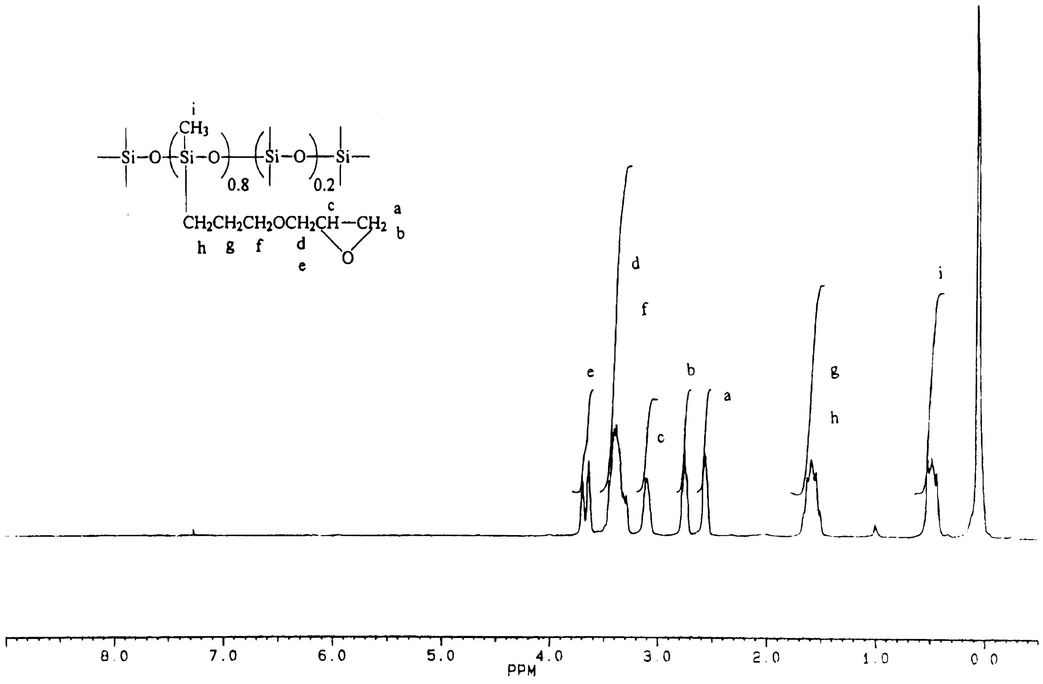


Fig. 1. ^{1}H NMR spectrum of the SG copolymer.

equation:

$$\mathrm{EEW} = \frac{W \times Nv \times 1000}{(B - A) \times F}$$

where W is the weight of SG-copolymer used; Nv the sample residue remaining (wt%) after heating in an oven at 150°C for 30 min, B the NaOH solution used in the blank solution, A the NaOH solution used in the sample solution and F the concentration of the NaOH solution.

2.4. Methods and measurements

^{1}H NMR spectra were recorded on a 200 MHz instrument (Bruker AC200) using $CDCl_3$ as solvent and tetramethylsilane as internal standard. Molecular weight of SG copolymer was determined by gel permeation chromatography (Waters 510).

Differential scanning calorimetry (DSC) (Perkin–Elmer DSC7) was used for characterizing the curing behaviors of DGEBA/SG blends. The sample used for DSC analysis was prepared by mixing SG copolymer, DGEBA and stoichiometric amounts of DICY with or without BDMA (an accelerator) with a miller. Subsequently, small samples (about 5 mg) were sealed in aluminum sample pans. DSC scans of curing reactions were conducted under a dry nitrogen atmosphere at a heating rate of 10°C/min.

The samples characterized with DMA and TGA were prepared by the following procedure: DGEBA, SG copolymer, DICY and BDMA (1.0% of total weight) were mixed well with a miller to give the required formulation. The mixture was degassed in a vacuum oven to remove any trapped air. The blends were pre-cured at 140°C for 1 h, and then cured at 160°C for 4 h.

Dynamic mechanical thermal analysis (DMA) (Perkin–Elmer DMA7) was performed using a three-point bending mode over a temperature range of 25–180°C at a heating rate of 10°C/min and a frequency of 1 Hz.

Thermal stability were determined with a thermogravimetric analyzer (TGA) (Perkin–Elmer TGA7) over a temperature range of 30–800°C at a heating rate of 20°C/min.

Scanning electron microscope (SEM) (JEOL JSM-35) was employed to examine the morphology of the surfaces of fractured samples.

3. Results and discussion

3.1. Synthesis and characterization of epoxy–siloxane copolymer (SG copolymer)

The epoxy–siloxane copolymer (SG copolymer) which contains epoxide groups on the side-chain was synthesized from the hydrosilylation reaction of poly(methylhydrosiloxane) copolymer (SH) and allyl glycidyl ether (AGE) as shown in Scheme 1. The SH copolymer was obtained by the acid-catalyzed equilibration of D4 and DC-1107 with hexamethyldisiloxane as an end-capper (Eq. (1)). The SH copolymer was then hydrosilated with allyl glycidyl ether in the presence of a Pt-catalyst to give the epoxy–siloxane copolymer (Eq. (2)). The ^{1}H NMR spectrum of the SG copolymer (Fig. 1) shows the complete disappearance of

Table 1
Curing characteristics of the blends of DGEBA and SG copolymer cured by DICY without accelerator and with accelerator (values in parenthesis)

Sample name	Content of SG copolymer (wt%)[a]	T_i (°C)[b]	T_p (°C)[c]	T_f (°C)[d]	Curing range (°C)
Neat DGEBA	0	162.5	197.5	244.7	82.4
		(97.5)	(145.6)	(186.0)	(88.5)
SG-0.1	9.1	161.2	199.4	242.8	81.6
		(102.0)	(146.1)	(188.8)	(86.8)
SG-0.4	28.6	165.1	199.2	234.2	69.1
		(93.8)	(142.1)	(190.6)	(96.8)
SG-0.7	41.2	169.1	200.4	247.4	78.3
		(91.7)	(136.7)	(209.6)	(117.9)
SG-1.0	50.0	180.2	202.6	232.9	52.7
		(103.2)	(152.0)	(206.4)	(103.2)
Neat SG	100	185.6	212.1[e]	243.9	58.3
		(127.4)	(164.8)	(244.9)	(117.5)

[a] The wt% of SG-copolymer content are calculated from the ratio of SG copolymer to the total weight of DGEBA and SG copolymer.
[b] T_i is the initial temperature of the curing reaction.
[c] T_p is the peak temperature during the curing process.
[d] T_f is the completed curing temperature.
[e] There are two exothermic temperatures (208.3 and 212.1°C) for the neat DGEBA curing process.

the Si–H resonance ($\delta = 4.7$ ppm) and the concomitant disappearance of the allyl group in allyl glycidyl ether ($\delta = 5.5$ and 6.0 ppm). In addition, the peaks at $\delta = 0.5$, 1.5 and 3.4 ppm are the resonance peaks of the silypropyl group, while those at $\delta = 2.5$, 2.7 and 3.1 ppm represent the resonance peaks of the epoxide group. These results clearly indicate that the hydrosilyation reaction was successful.

The mole content of the epoxy–siloxane unit in the copolymer can be obtained from the EEW titration. The EEW value of the SG copolymer is 190, so the mole fraction of the epoxy–siloxane unit in the SG copolymer can be calculated from the following equation:

$$\frac{1}{1 + (\mathrm{EEW} - 173)/74}$$

where 173 and 74 are the unit weights of the epoxy–siloxane unit and the dimethylsiloxane unit, respectively. Thus, the mole fraction of the epoxy–siloxane unit in the SG copolymer is calculated to be 81%. The number average molecular weight ($\bar{M}_n$) of the SG copolymer is 1.5×10^4 which was determined by gel permeation chromatography (GPC), using polystyrene as standards.

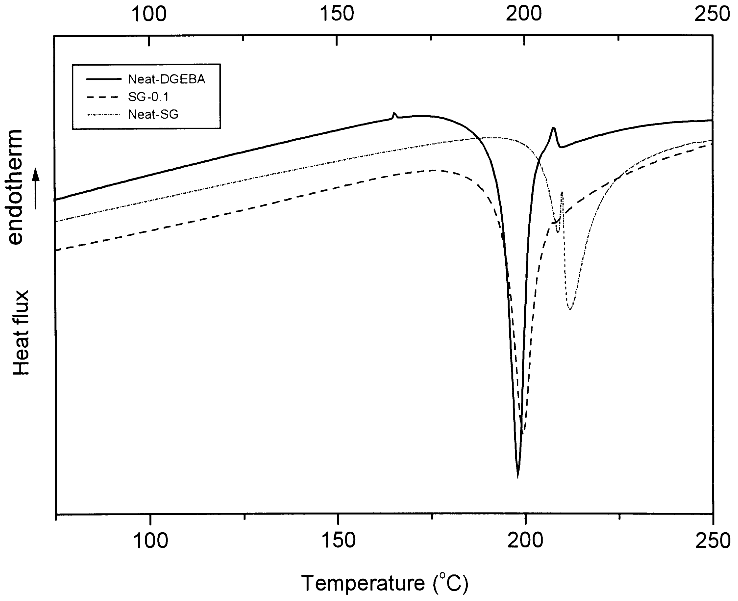


Fig. 2. Typical DSC measurements of the SG/DGEBA mixtures without addition of BDMA.

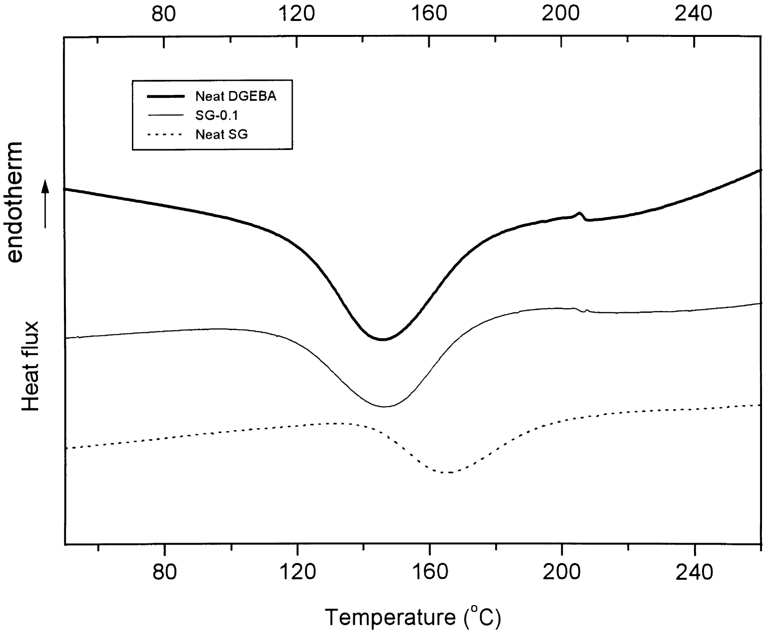


Fig. 3. Typical dynamic DSC measurements of the SG/DGEBA mixtures prepared with addition of BDMA.

3.2. Curing behavior of SG copolymer and DGEBA

In the present study, SG copolymer and DGEBA were blended at different ratios (see Table 1) and dicyandiamide (DICY) was used as a curing agent. Because its latent temperature can be up to 150°C, DICY has been widely used as a curing agent for epoxy resins in adhesive, composites, printed circuit boards and powder coatings applications [18]. However, DICY is a solid with high melting point (m.p. 213°C, determined by DSC), and is nearly insoluble in the SG copolymer and DGEBA blends. Therefore, the resins were blended with DICY by a miller to make an extremely micronized DICY to obtain a homogeneous system ensuring a nearly complete curing reaction.

From its chemical structure, DICY is expected to have a functionality of four due to the presence of four amino groups. Therefore, the stoichiometry for the epoxy–DICY reaction was selected to be 4:1 in this study, although some investigations [19] show that DICY has functionality greater than four toward epoxy resins. Since the curing temperature of DICY to epoxy resins is about 180°C, and for comparison, benzyldimethylamine (BDMA) is also used as a catalyst to accelerate the curing reaction and to reduce the curing temperature.

Fig. 2 shows the typical curing characteristics of SG/DGEBA mixtures as a function of curing temperature without adding any accelerator. The curing behavior results for a series of DGEBA epoxies with increasing amounts of added SG copolymer are summarized in Table 1. In Fig. 2, a single exothermic peak was observed for the neat DGEBA curing process and a small endothermic peak near 210°C corresponded to the melting peak of the remaining unreacted

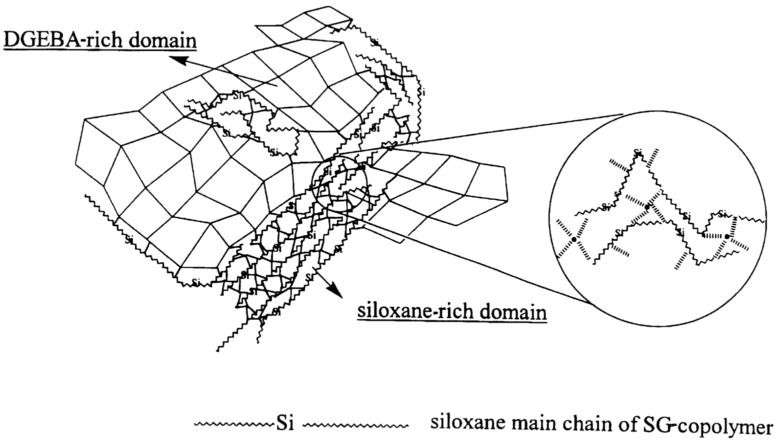
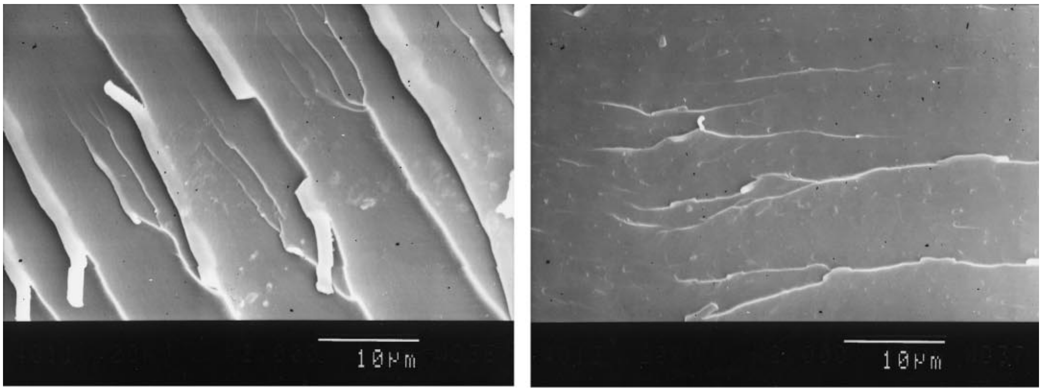
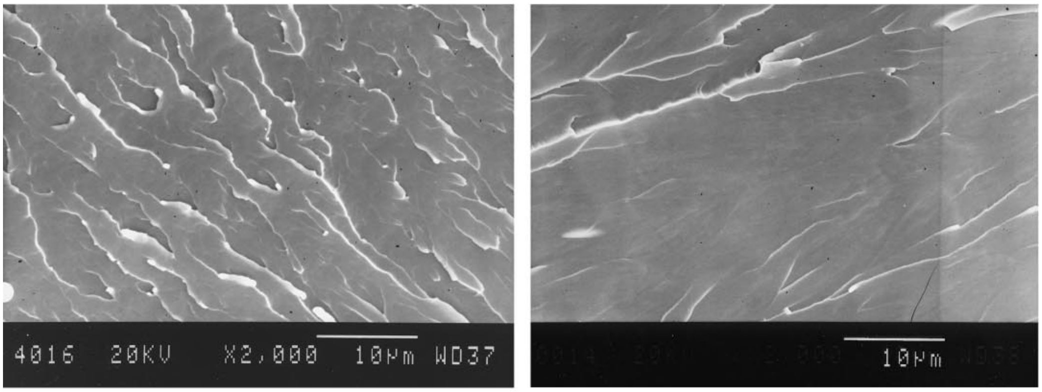


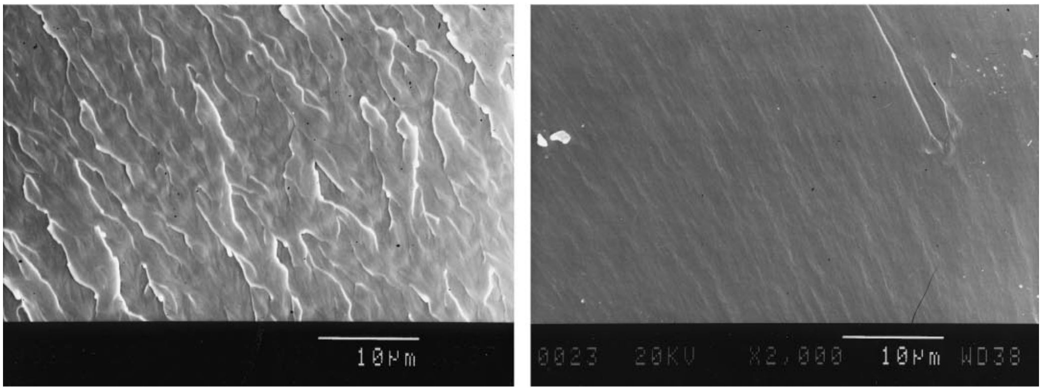
Fig. 4. Proposed morphology of the cured SG/DGEBA blends.



(a) Neat Epoxy (b) SG-0.1



(c) SG-0.4 (d) SG-0.7



(e) SG-1.0 (f) Neat SG

Fig. 5. SEM micrographs of the fracture surfaces of cured SG/DGEBA blends.

DICY. On the other hand, for neat SG copolymer, the highest initial reaction temperature (T_i, 185.6°C) and two distinct exothermic peaks (208.3 and 212.1°C) were observed. This indicates that the curing reaction for neat SG copolymer was not remarkable until DICY began to melt and the reaction became fast after DICY completely melted; hence, a greater reaction heat was released, resulting in a larger exothermic peak. For the blends of SG copolymer and DGEBA, only one exothermic peak was observed, and the peak temperature (T_p) increased with an increase in the siloxane content (Table 1) due to the lower reactivity of the SG copolymer. In addition, Fig. 3 gives the typical curing characteristics of SG/DGEBA mixtures when adding BDMA as an accelerator; the detailed results are shown in parenthesis in Table 1. Comparing these results to the curing behavior of the blend series obtained without adding BDMA, it was found that same trend was obtained for the T_i, T_p and T_f values; i.e. they increased with increasing siloxane content. The higher T_i, T_p and T_f temperature for the SG/DGEBA mixtures can be attributed to the necessity for a higher reaction temperature required for the large molecules (SG copolymer). Further, the T_i, T_p and T_f temperatures were lower than those obtained without adding BDMA by 60–70°C, indicating that BDMA

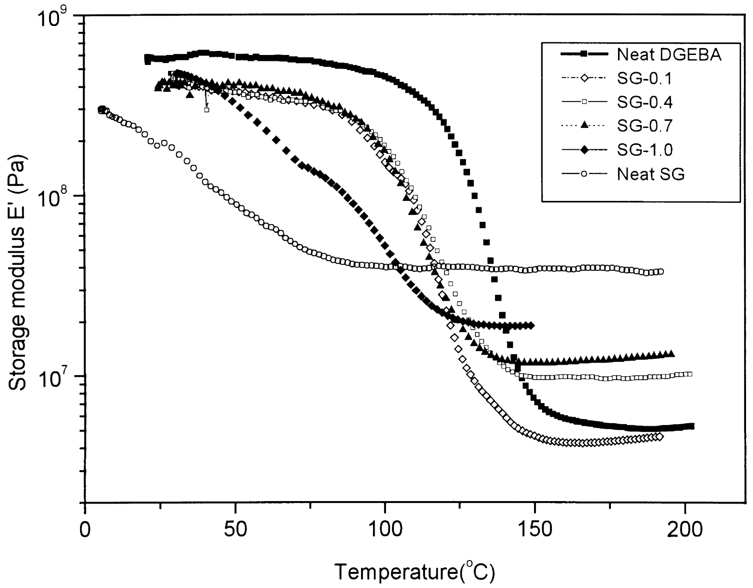


Fig. 6. Storage modulus of the cured SG/DGEBA blends.

can effectively accelerate the curing reactions of SG/DGEBA blends.

From Table 1, it is interesting to see that the trend for the curing temperature range ($T_f - T_i$) for the system without adding BDMA is opposite to that observed for the system added BDMA, i.e. in the case where BDMA was absent, a narrower curing range is obtained with increasing siloxane content, but with additional BDMA as a accelerator, the curing range increased with an increase in the SG copolymer content. This can be explained by the following. In the system without adding BDMA, addition of more SG copolymer results in higher T_is due to its lower reactivity, but T_fs are nearly the same whatever the amount of SG copolymer added into the curing system. As a result, a narrower curing range is found with increasing siloxane content. Nevertheless, in the system with additional BDMA incorporated, T_i and T_f both decreased than that in the case without adding BDMA. This means that the more amount of SG copolymer, compared to the system without adding BDMA, may react with itself or with DGEBA to form a continuous matrix in the early stage of curing reaction. Also, a siloxane-rich domain and a DGEBA-rich

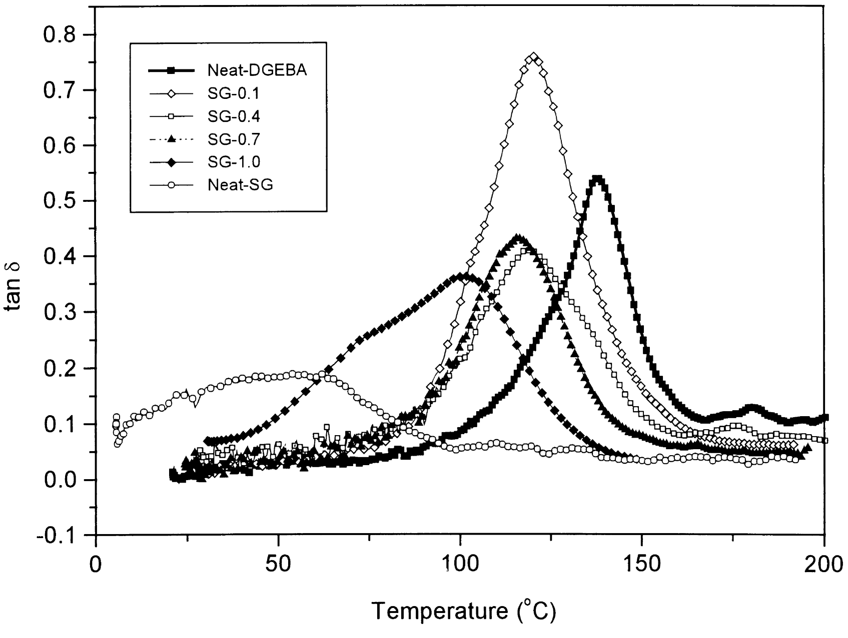


Fig. 7. tan δ relaxations of the cured SG/DGEBA blends.

Table 2
DMA and TGA characteristics of blends of DGEBA and SG copolymer cured using DICY with BDMA

Sample name	E'_g (Pa)	E'_r[a] (Pa)	$(\tan \delta)_{max}$	T_g (°C)	T_d[b] (°C)	R_{800}[c] (%)
Neat DGEBA	5.0×10^8	5.5×10^6	0.54	137.5	376.6	0.32
SG-0.1	3.8×10^8	4.6×10^6	0.76	120.1	377.3	13.2
SG-0.4	3.7×10^8	9.8×10^6	0.41	118.5	379.5	17.7
SG-0.7	4.0×10^8	1.2×10^7	0.43	116.5	383.2	18.7
SG-1.0	3.2×10^8	1.9×10^7	0.36	100.5	387.2	18.4
Neat SG	(9.0×10^7)[d]	4.3×10^7	0.19	54.2	394.8	23.4

[a] The E'_r value is taken at T_g + 30°C.
[b] Temperature of 10% weight loss.
[c] Residual wt% at 800°C.
[d] For neat SG, there is no distinct glassy state, this value is taken at 0°C.

domain may possibly form and chemically bond in the continuous matrix (see Fig. 4) due to the incompatibility and the difference in reactivity between SG copolymer (slower) and DGEBA (faster). Thus, a higher crosslinking density network formed by SG copolymer may reduce the mobility of the half-cured blended network and result in a wider curing range.

3.3. Morphology of cured blends

The SEM micrographs of cold-fracture surfaces (unetched) of a series of cured blends (DGEBA + SG copolymer + DICY + BDMA) are shown in Fig. 5(a)–(f). In Fig. 5(a), the cured neat DGEBA network exhibits a very smooth surface. As the content of SG copolymer in the network increases, the fracture surfaces become more rough and exhibit more irregular stripes in the matrix, Fig. 5(b)–(e). The striped and rough morphology of these blends reflects the extent of incompatibility between DGEBA and SG copolymer in the cured blends. Even though a distinct phase separation in the DGEBA–SG matrix was not observed from SEM micrographs, SG-1.0 does exhibit some degree of phase separation as observed from the loss spectra. It is clear that the siloxane-rich and DGEBA-rich domains (Fig. 4) exist separately in the blended networks.

3.4. Dynamic viscoelastic analysis

Most polymeric materials can be considered to be viscoelastic solids that exhibit both viscous and elastic properties. Dynamic mechanical thermal analysis (DMA) is an efficient tool to measure the viscoelastic properties and obtain information about the microstructure of crosslinked networks. The storage modulus (E') and the loss factor (tan δ) for a series of mixtures of SG copolymer and DGEBA cured upon DICY with BDMA as a accelerator are shown as a function of temperature in Figs. 6 and 7. From the viscoelastic spectra, several parameters can be determined: the glassy modulus (E'_g), rubbery modulus (E'_r, taken at T_g + 30°C), the glass transition temperature (determined by the tan δ peak) and the amplitude of the damping peak $(\tan \delta)_{max}$; these values are tabulated in Table 2.

It can be seen that E'_g of these cured networks drop from 5×10^8 to 3×10^8 Pa (Fig. 6) when adding SG copolymer into the blends. Moreover, when the amount of SG copolymer was increased up to 50 wt% (SG-1.0), the temperature range of glassy state became very short compared to the networks formed by the neat DGEBA, SG-0.1, -0.4 and -0.7, and the neat SG network exhibited no glassy state even the temperature was down to about 0°C. These observations can be explained by the flexibility of the siloxane bonds in the SG copolymer, which decreases the stiffness of the crosslinked networks.

From the rubber elasticity theory [20], it may be assumed that the effect of internal energy of the formed network can be neglected above T_g and by relating the changes in entropy, due to the motion of segments among the network strands. Thus, the variation in the rubbery modulus (E'_r) can be expressed as a function of crosslinking density by the following relationship:

$$E'_r = 3\theta\rho \mathrm{R}T/M_c$$

where R is the gas constant, T the absolute temperature, ρ the density of the crosslinked network, θ is the 'front factor' (the value of θ is close to unity), and M_c the average molecular weight between crosslinks. The above equation shows that E'_r is proportional to $1/M_c$; in other words, an increase in the crosslinking density will cause an increase in E'_r.

Consequently, in the cured blends of SG copolymer and DGEBA, there is an observed trend that E'_r increases with increasing amounts of SG copolymer added (Table 2), despite the observation that E'_r of SG-0.1 (with 9 wt% of SG copolymer) and that of the neat DGEBA are nearly the same. This result shows that the crosslinking density of the SG copolymer network is higher (or M_c is smaller) than that of DGEBA. As mentioned earlier in the discussion of blend morphology, the resulting cured networks are not homogeneous so that two types of domains form in the networks, i.e. DGEBA-rich and siloxane-rich domains as demonstrated in Fig. 4. It is obvious that the average molecular weight between crosslinks formed by SG copolymer is smaller than that obtained from DGEBA. Since the epoxide groups in the SG copolymer are attached on the side chain (with a

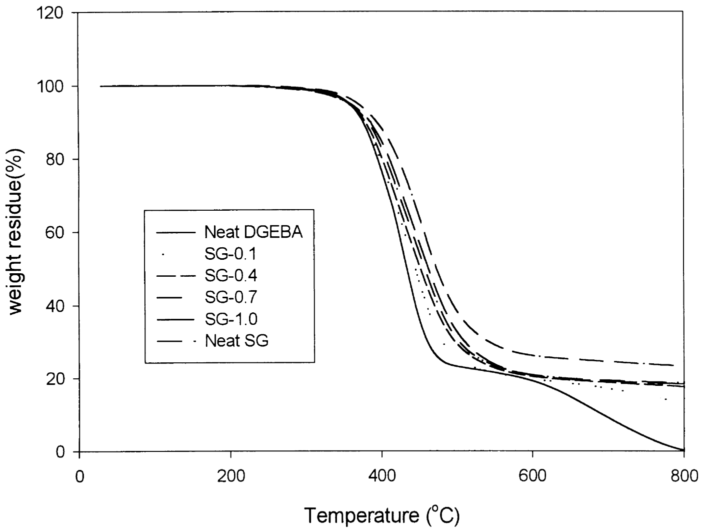


Fig. 8. TGA thermograms of the cured SG/DGEBA blends.

mole fraction of 81%) compared to the case of DGEBA where the epoxide groups are on the terminal ends of the molecule, the average distance between the two crosslinking sites is shorter and which results in a higher crosslinking density (see Fig. 4). As a result, the increasing concentrations of SG copolymer present in the cured networks leads to the presence of the larger and more siloxane-rich domains in the blend, resulting in higher values of E'_r.

Further, the analysis of the shape of the loss spectra (tan δ versus temperature, Fig. 7) can provide additional information about the structure of the crosslinked networks. Thus, the height of the α-transition (or glass transition temperature), (tan $\delta)_{max}$, for a thermosetting mixture supplies information about the extent of cure, i.e. (tan $\delta)_{max}$ becomes lower as the distance between crosslinks decreases [15,21,22]. As seen in Table 2, the (tan $\delta)_{max}$ for neat DGEBA decreases with increasing amounts of SG copolymer in the crosslinked networks, except that (tan $\delta)_{max}$ of SG-0.1 is significantly higher than those of the others. The result is consistent with the trend obtained from the E'_r-determination; i.e. the higher siloxane content will lead to a more densely crosslinked network.

On the other hand, the widths at the half-height on the α-relaxation peak are sensitive to the homogeneity of the networks [15,21,22]. In Fig. 7, it is clear that neat DGEBA, SG-0.4 and SG-0.7 exhibit a single glass transition peak, while SG-1.0 has a distinct shoulder peak near 75°C. This means that the siloxane-rich domains do not separate from the whole continuous phase of the SG DGEBA networks until the SG copolymer content exceeds 50 wt%. Besides, due to the incompatibility between DICY and SG copolymer, the network obtained from neat SG is not as homogeneous as expected, so the tan δ peak is broad and covers a wide temperature range.

It is interesting to note that SG-0.1 is an exception to the E'_r and (tan $\delta)_{max}$ trend, i.e. it has the lowest E'_r value and the highest (tan $\delta)_{max}$ in the series of blends. This probably arises from small amounts of long, soft siloxane chains which exist in the DGEBA network which results in a more loosely crosslinked network compared to the network formed by only neat DGEBA. At the same time, in the SG-0.1 network, the siloxane content is too low to form a large siloxane-rich domain and the siloxane chains are finely dispersed in the continuous DGEBA matrix. In addition, there is a small shoulder at around 110°C in the tan δ peak for SG-0.1, which indicates the chain movements in networks become easier due to the flexibility of the –Si–O–Si–O– bonds if a small portion of the polymer chains in the neat DGEBA crosslinked network are replaced by the siloxane segments. In contrast, if too many and too large siloxane-rich domains are formed by adding too much SG copolymer into the blends, the crosslinking density of the cured blends are too high to promote the effective toughening of the cured blends.

To summarize, the SG-0.1 network cured using DICY with BDMA as a catalyst has a better capability to damp out noise and vibration, which are particular forms of dynamic mechanical motion.

3.5. Thermal stability of cured epoxy mixture

The thermal stability of the series of the cured blends (DGEBA + SG copolymer + DICY + BDMA) was investigated by TGA, and the results are shown in Fig. 8 and Table 2. The thermograms show that there is only one decomposition stage in the heating process. The temperature for 10% weight loss (T_d) of the cured neat DGEBA network is 376.6°C, and the T_d increases as the concentration of SG copolymer increases (Table 2). This indicates that the thermal stability in the blend series improves as a greater amount of siloxane components are incorporated into the cured networks. The thermograms also show that the residual weight percent at 800°C (R_{800}) of the cured network for neat epoxy, SG-0.1, −0.4, −0.7 −1.0 and neat SG copolymer are 0.3, 13.2, 17.7, 18.7, 18.4 and 23.4%, respectively; i.e. the R_{800} value increases upon addition of increasing amounts of SG copolymer. Obviously, the thermal stability and the residual weight are improved when siloxane segments are introduced into the cured blends.

4. Conclusions

In this study, epoxy–siloxane copolymers (SG copolymer) which contain epoxide groups on the copolymer side chain were successfully synthesized using the hydrosilylation reaction. The SG copolymer was then used to modify the toughness of a neat epoxy resin (DGEBA). The SG copolymer and DGEBA form a clear solution when they are blended together at room temperature which shows that the compatibility between the two polymers is very good. However, from the DSC, SEM and DMA results, for a matrix of cured SG/DGEBA blends using DICY as the curing agent and using BDMA as a accelerator, there exist siloxane-rich and DGEBA-rich domains, since the differences in reactivities for each material to the curing agent will result in phase separation in the cured matrix. From the viscoelastic spectra, the cured SG-0.1 network has the best capability to toughen the epoxy resin because the siloxane chains are finely dispersed in the continuous cured DGEBA matrix. Finally, the thermal stabilities of the cured SG/DGEBA mixtures were investigated by TGA; the results indicate that the thermal stability and the residual weight were improved when siloxane segments are incorporated into the epoxy resin.

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