Polymer Vol. 36 No. 25, pp. 4753–4760, 1995

0032-3861/95/\$10.00+0.00

# Effect of curing agent on the phase behaviour of epoxy resin/phenoxy blends

**Qipeng Guo**
*Department of Material Science and Engineering, University of Science and Technology of China, Hefei 230026, People's Republic of China*
*(Received 21 December 1994; revised 5 April 1995)*

The poly(hydroxyether of bisphenol A) (phenoxy) was found to be miscible with uncured bisphenol-A-type epoxy resin, i.e. the diglycidyl ether of bisphenol A (DGEBA), as shown by the existence of a single glass transition temperature ($T_g$) over the whole composition range. However, dynamic mechanical analysis (d.m.a.) and scanning electron microscopy (SEM) studies revealed that the phenoxy exhibits different miscibilities with five cured epoxy resins (ERs). The miscibility and phase behaviour, as well as the resulting morphology of the cured blends, are dependent on the choice of cure agent. For the blends cured with 4,4′-diaminodiphenylmethane (DDM) and aliphatic anhydrides, i.e. maleic anhydride (MA) and hexahydrophthalic anhydride (HHPA), no phase separation, as indicated by both d.m.a. or SEM. However, for the blends cured with 4,4′-diaminodiphenyl sulfone (DDS) and phthalic anhydride (PA), both d.m.a. and SEM studies clearly show evidence of phase separation. The SEM study shows that two phases interact well in the PA-cured blends while the interface bonding between the two phases is relatively poor in the DDS-cured blends. The phenomena have been discussed from the points of view of both thermodynamics and kinetics.

**(Keywords: epoxy resin; phenoxy; phase behaviour)**

## INTRODUCTION

Epoxy resins are a major class of thermosetting polymers which are widely employed as matrices for composite materials and as structural adhesives[1–4]. They are amorphous, highly crosslinked polymers and this structure results in these materials possessing a high modulus and fracture strength, low creep, and good performance at elevated temperatures. However, it also leads to a low toughness and poor crack resistance. One of the most successful methods of increasing their toughness is to incorporate a second phase of dispersed rubbery particles into the crosslinked polymer[5–11]. The addition of rubbery materials to epoxy resins has been shown to enhance their fracture toughness, while at the same time resulting in a lowering of their glass transition temperatures ($T_g$s) and thermal and oxidative stability. Recently, engineering thermoplastics have been also used to toughen epoxy resins[12–18]. Owing to the high moduli and high $T_g$s of these thermoplastics, values for the modulus and $T_g$ of the modified epoxy resin can reach or even surpass those of the pure resin.

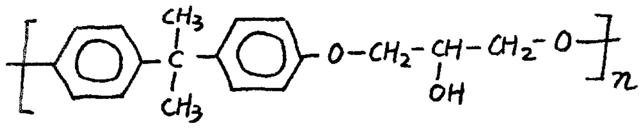
However, only a few systematic studies have been carried out on the overall compatibility and phase separation in blends of thermoset resins with elastomers or thermoplastics[13,19–27]. Since the resulting morphology and extent of phase separation is known to affect the optical and mechanical properties of the cured blends, the need for an understanding of the phase separation processes taking part in thermosetting blends is of great practical importance.

On the other hand, it is also of academic interest to examine the phase behaviour of such crosslinked polymer mixtures, i.e. interlocked systems containing one crosslinked and one linear component, which are usually known as semi-interpenetrating polymer networks (semi-IPNs) in the highly crosslinked cases. From the point of view of thermodynamics, an increase in the molecular weight for either of the components of miscible blend would decrease the cloud-point temperature. Therefore, phase separation as a result of crosslinking should be expected. Indeed, we have found that phase separation occurs as the crosslinking process proceeds in our earlier examination of miscible blends of uncured epoxy resin and poly(ethylene oxide) (PEO)[28,29]. However, the complex interrelationship between morphology and composition has not yet been clarified and is currently the subject of a research programme taking place in our laboratory.

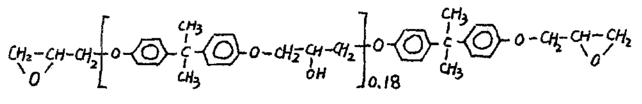
In our previous work[30,31], we have studied the morphology and phase behaviour of cured epoxy resin containing phenolphthalein poly(ether ether ketone) (PEK-C). The latter was proved to be miscible with the poly(hydroxyether of bisphenol A) (phenoxy) which may be considered to be a model epoxy resin[32,33]. The miscibility of PEK-C with uncured bisphenol-A-type epoxy resin, i.e. the diglycidyl ether of bisphenol A (DGEBA), was established as expected[30,34]. However, it has been shown that the cured epoxy resin/PEK-C blends are all incompatible and have a two-phase structure[30,31]. Furthermore, our recent study[35] shows that the overall compatibility and the resulting morphology of the cured blends are dependent on the choice of cure agent in the cured blends of epoxy resin with phenolphthalein poly(ether ether sulfone) (PES-C), although PES-C has been shown to be miscible with DGEBA. It was found that PES-C exhibits more overall compatibility with the amine-cured epoxy resins than with the anhydride-cured resins, which has been considered to be due mainly to the presence of hydroxyl groups in the amine-cured system, which offer an excellent potential for hydrogen-bonding interactions with the ester side groups and/or the ether oxygens of the PES-C in the blends. Similarly, it has been shown by other authors[36,37] that poly($\epsilon$-caprolactone) (PCL) exhibits more miscibility with amine-cured epoxy resins than with anhydride-cured resins because of the existence of opportunities for hydrogen bonding in amine-cured epoxy resin/PCL blends.

In this paper, we further report the results of work on the blends of phenoxy with a bisphenol-A-type epoxy resin, i.e. DGEBA. In particular, attention is paid to the effect of crosslinking and the choice of cure agent on the overall compatibility and the resulting morphology of the cured blends. Five cure agents have been used in this study. The structures of phenoxy and DGEBA are as follows:

Poly(hydroxyether of bisphenol A) (phenoxy)



Diglycidyl ether of bisphenol A (DGEBA)



This system was chosen for three reasons. First, as has been noted above, phenoxy can be considered as being a model epoxy resin, i.e. it is the linear high-molecular-weight polymer of DGEBA. Therefore, both the crosslinked and linear components in the blends will have the same structural units appearing in the chains. Secondly, PEO, PEK-C, PES-C and PCL, previously used as linear components of the ER blends[28–31,35–37], are all proton-accepting polymers which offer an excellent potential for forming hydrogen bonds in the amine-cured blends. As a result, it has been observed that PCL and PES-C exhibit more miscibility with amine-cured epoxy resins than with anhydride-cured resins. In addition, phenoxy is a proton-donating polymer and so offers opportunities for hydrogen bonding with the anhydride-cured epoxy resins, rather than with the amine-cured resins, and this will enhance the miscibility of the anhydride-cured blends. Thirdly, the pendant hydroxyl groups of phenoxy may become involved in the cure reaction with anhydride, thus resulting in interchain crosslinking, with the formation of copolymers acting as a compatabilizer for the anhydride-cured blends.

## EXPERIMENTAL

### *Materials*

The phenoxy resin, having a quoted average molecular weight $\bar{M}_w$ of 34 000, was supplied by Scientific Polymer Products, Inc., Japan. The diglycidyl ether of bisphenol A (DGEBA), E-51, with an epoxy equivalent of 185–210, was supplied by Wuxi Resin Works, Jiangsu, China. The hardeners used were 4,4′-diaminodiphenylmethane (DDM) (purity > 98%), 4-4′-diaminodiphenyl sulfone (DDS) (purity > 98%), maleic anhydride (MA) (purity > 98), phthalic anhydride (PA) (purity > 98%), and hexahydrophthalic anhydride (HHPA) (purity > 98%). The solvents used here were Analar grade chloroform and tetrahydrofuran (THF).

### *Preparation of samples*

Blends of phenoxy with DGEBA were prepared by solution casting from chloroform. The solvent was allowed to evaporate slowly at room temperature, and in order to remove the residual solvent, the blend was then dried in a vacuum oven at 50°C for 2 weeks.

The ER/phenoxy blends were prepared by mixing preweighed phenoxy, DGEBA and hardener in the smallest possible amount of THF. The solution was then cast on to an aluminium plate to form the film specimens, and residual solvent removed under vacuum at room temperature. The blend films thus obtained were then cured successively at 150°C for 8 h, 200°C for 4 h and finally at 250°C for 4 h.

### *Differential scanning calorimetry (d.s.c.)*

The glass transition temperature ($T_g$) of the phenoxy/DGEBA blends was determined with a Perkin–Elmer DSC-7 differential scanning calorimeter at a heating rate of 20°C min$^{-1}$. The midpoint of the change in slope of the heat capacity plot of the second scan was taken as the value of the $T_g$.

### *Dynamic mechanical measurements*

Dynamic mechanical measurements were carried out on a Rheovibron Model DDV II dynamic viscoelastometer (Toyo Baldwin Co., Japan); the frequency used was 3.5 Hz, with a heating rate of 3.0°C min$^{-1}$. The sample dimensions were $4.0 \times 0.4 \times 0.04\,\mathrm{cm}^3$.

### *Morphological observations*

The specimens were fractured under cryogenic conditions using liquid nitrogen. The fractured surface was then immersed in chloroform at room temperature for 15 h. The phenoxy phase and/or the sol fraction of the epoxy network were preferentially etched by the solvent while the cured ER phase remained relatively little unaffected. The etched samples were then dried to remove the solvent. A JEOL JXA-840 scanning electron microscope was used for the observations, prior to which the surfaces were coated with thin layers of platinum of ~ 200 Å in thickness.

## RESULTS

### *Phenoxy/DGEBA blends*

Blends containing phenoxy are among the most studied of such polymer systems. Harris *et al.*[38] have described numerous miscible and immiscible blends of this polymer with a series of aliphatic and aromatic polyesters. Robeson and coworkers have reported miscible blends of this polymer with poly(butylene terephthalate)[39] and polyethers[32], such as PEO and poly(vinyl methyl ether). Equiazabal *et al.*[40] have

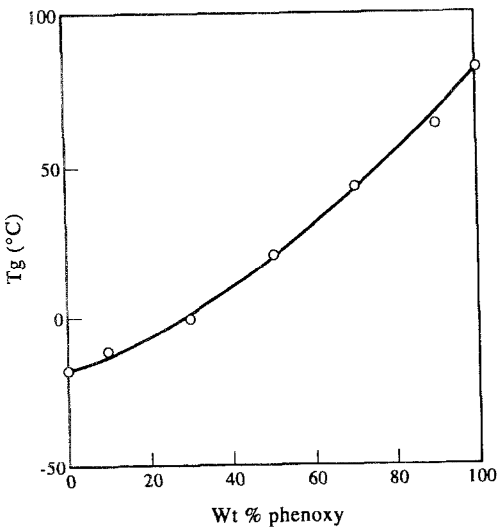


**Figure 1** Composition dependence of the glass transition temperature of DGEBA/phenoxy blends; the curve is as predicted by the Gordon–Taylor equation using a $k$ value of 0.62

established the miscibility of phenoxy with poly($N$-vinyl-2-pyrrolidone). In all cases, the miscibility observed has been attributed to the intermolecular interactions between the two components of the blend. These interactions could arise from possible hydrogen bonding involving the pendant hydroxyl groups of the phenoxy. Direct evidence of such a hydrogen-bonding interaction has been found in the results from the Fourier transform infra-red spectroscopy studies reported by Moskala and Coleman[41].

In this present case, all of the phenoxy/DGEBA blends were transparent, and no phase separation occurred when heating up to ~ 280°C. A d.s.c. study showed that each blend has a single composition-dependent $T_g$, thus indicating its single-phase nature. Therefore, phenoxy is miscible with DGEBA over the entire composition range. *Figure 1* summarizes the $T_g$ values obtained using d.s.c., as a function of the blend composition.

Several theoretical and empirical equations have been used to describe the $T_g$–composition dependence of miscible blends. One of these, the Gordon–Taylor equation[42], is written as follows:

$$T_g = (W_1 T_{g_1} + k W_2 T_{g_2})/(W_1 + k W_2) \qquad (1)$$

where $T_g$ is the glass transition temperature of the blend, with $T_{g_1}$ and $T_{g_2}$ being the glass transition temperatures of components 1 and 2, respectively; $W$ is the weight fraction, and $k$ is a constant. The curve in *Figure 1* is drawn using the Gordon–Taylor equation with a $k$ value of 0.62, and fits the experimental data well.

Prud'homme and coworkers have suggested[43,44] that the $k$ value can be taken as a semiquantitative measure of the strength of interaction between the components of the blend. For instance, in blends of poly(ε-caprolactone) with chlorinated polyethylene, poly(vinyl chloride) (PVC) and chlorinated PVC, $k$ increases from 0.26 to 1.0. When such an approach is used for the DGEBA/phenoxy blends, we note that $k$ (0.62) has only a moderate value, suggesting that the interaction between DGEBA and phenoxy is not strong. The miscibility of the DGEBA/phenoxy blends can be considered to be due primarily to a non-negligible entropy contribution, as the molecular weight of DGEBA is rather low. In addition, the epoxy groups of DGEBA can interact with the pendant hydroxyl groups of the phenoxy through hydrogen bonding, which is also responsible for the miscibility.

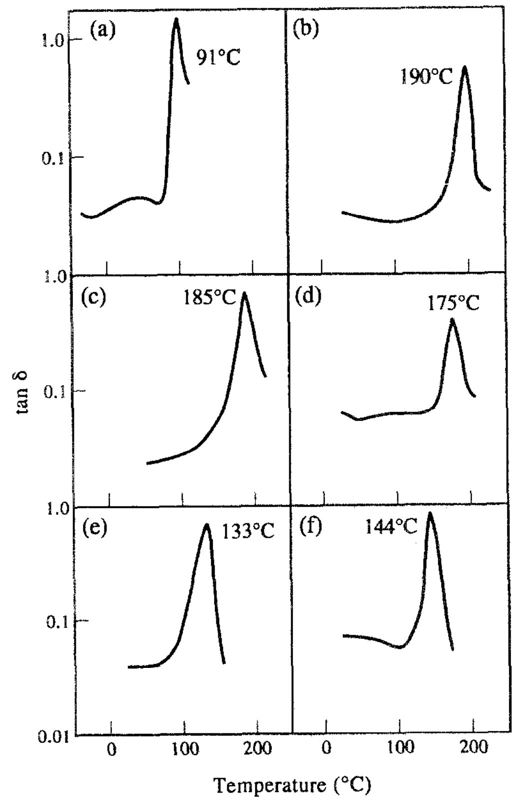


**Figure 2** Dynamic mechanical spectra of: (a) phenoxy; (b) 100 DGEBA, 25 DDM; (c) 100 DGEBA, 30 DDS; (d) 100 DGEBA, 50 MA; (e) 100 DGEBA, 75 PA; (f) 100 DGEBA, 80 HHPA

### *DDM-cured blends*

The dynamic mechanical spectrum of phenoxy is shown in *Figure 2a*. There exists a well defined relaxation peak at 91°C on the tan $\delta$ vs $T$ plot, corresponding to the glass–rubber transition of phenoxy.

All of the cured ER samples were transparent and their dynamic mechanical spectra are shown in *Figure 2*. They all clearly display a well defined relaxation peak on the tan $\delta$ vs. $T$ curves, which correspond to the $T_g$ values of the ER cured with 25 phr DDM (190°C), 30 phr DDS (185°C), 50 phr MA (175°C), 75 phr PA (133°C), and 80 phr HHPA (144°C), respectively (phr = parts per hundred parts of resin). It is noted that the $T_g$ value obtained for the DDS-cured ER is considerably lower than that documented in the literature (200–220°C)[45]. This is due to the approximate stoichiometric ratios used in this study.

The cured blend containing 100 DGEBA, 25 DDM and 25 phenoxy was transparent. *Figure 3* shows the dynamic mechanical spectrum of this blend. Only one glass transition peak is observed on the tan $\delta$ vs. $T$ curve and its value (174°C) is intermediate between that of the pure phenoxy (92°C) and the neat DDM-cured ER (190°C), suggesting that this blend had a homogeneous one-phase structure, and that phenoxy is miscible with DDM-cured ER.

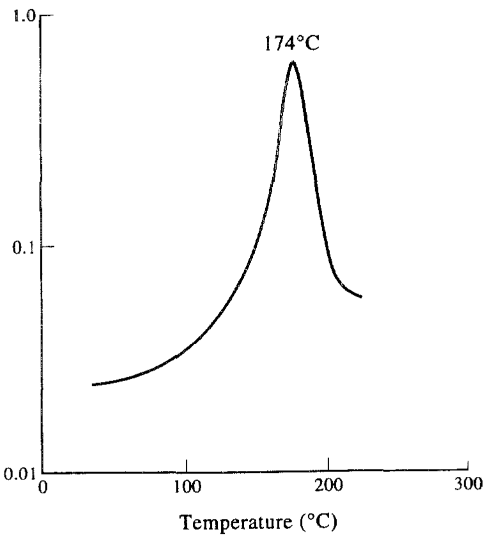


**Figure 3** Dynamic mechanical spectrum of the cured blend containing 100 DGEBA, 25 DDM, and 25 phenoxy

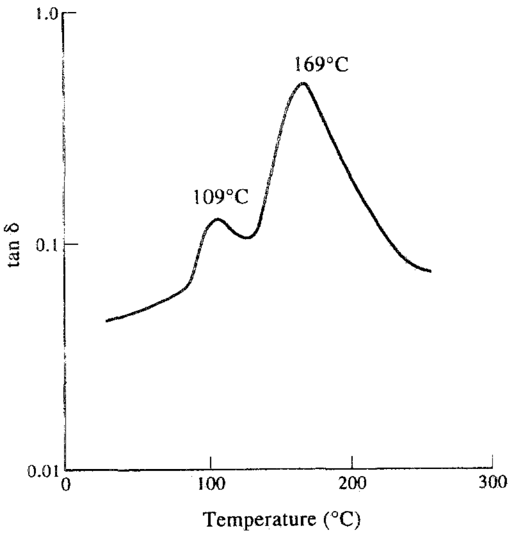


**Figure 5** Dynamic mechanical spectrum of the cured blend containing 100 DGEBA, 30 DDS, and 25 phenoxy

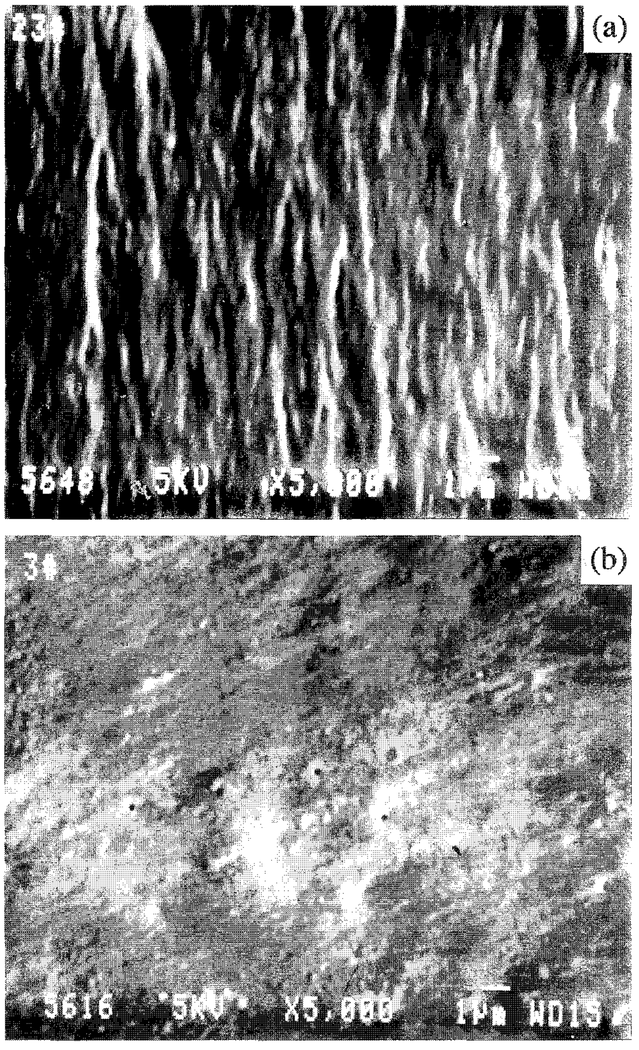


**Figure 4** Fracture surfaces of the cured blend containing 100 DGEBA, 25 DDM, and 25 phenoxy: (a) unetched; (b) etched in chloroform for 15 h

A scanning electron micrograph of the fractured surface of the DDM-cured ER/phenoxy blend (*Figure 4a*) appears to show no evidence that the blend is heterogeneous. To enable further examination to take place, the fractured surface of the specimen was etched to remove phenoxy, so that the morphology of the specimen may be more clearly shown. The scanning electron micrograph of the etched specimen given in *Figure 4b* displays no obvious characteristics of a phase-separated structure for an inhomogeneous blend. These observations support the results obtained by the d.m.a. study that the DDM-cured ER/phenoxy blend is homogeneous. It is also clear that phenoxy is completely miscible with DDM-cured ER.

### *DDS-cured blends*

The sample of the cured blend containing 100 DGEBA, 30 DDS and 25 phenoxy was opaque. *Figure 5* shows its dynamic mechanical spectrum which clearly displays two maxima on the $\tan\delta$ vs. $T$ curve, which correspond to the $T_g$ values of the phenoxy-rich phase (109°C) and the DDS-cured ER phase (169°C), respectively. It is evident that phase separation had occurred in the DDS-cured ER/phenoxy blend, and that phenoxy is not miscible with the DDS-cured ER. It can also be seen from the figure that the $T_g$ of the cured ER phase (169°C) is rather lower than that of the neat system (185°C). The lowering of the $T_g$ of the ER phase implies that there is some phenoxy dissolved in the cured ER phase. However, the $T_g$ value of the phenoxy-rich phase (109°C) is somewhat higher than that of pure phenoxy (91°C), which can be considered as a result of the crosslinking of ER in the phenoxy-rich phase. The d.m.a. study clearly shows that the DDS-cured ER/phenoxy blend is immiscible.

Scanning electron micrographs of both unetched and etched specimens clearly show that the blend had a two-phase structure and that domains of phenoxy-rich phase are dispersed in a continuous phase of DDS-cured ER (*Figure 6*). The size of the domains is in the range 0.2–5 µm in diameter. The interface between phenoxy and the DDS-cured ER is not strongly bonded. All of these

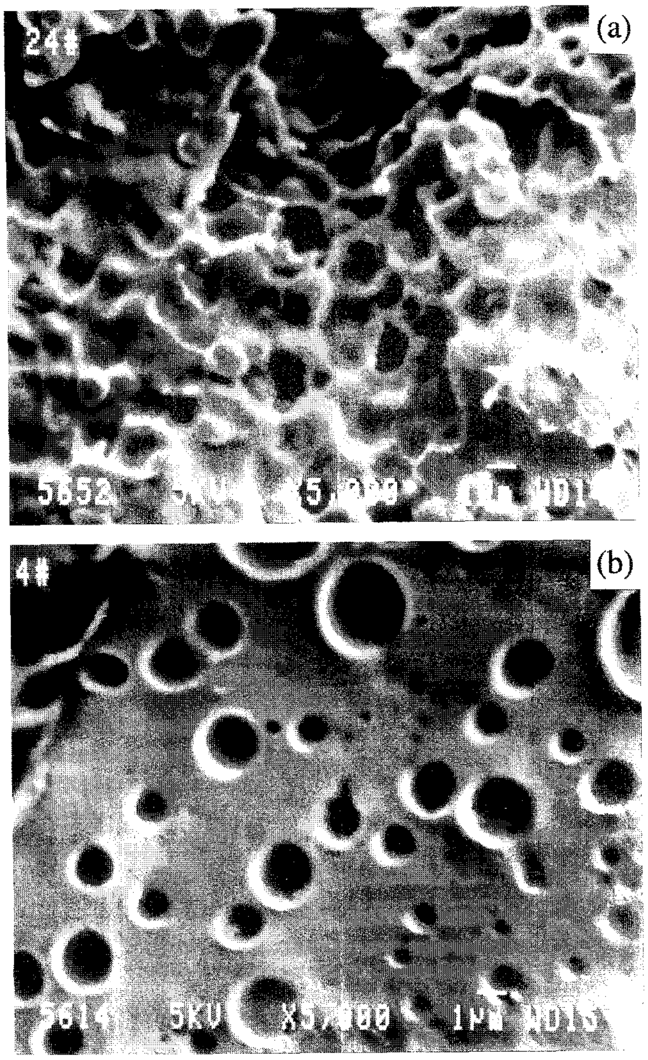


**Figure 6** Fracture surfaces of the cured blend containing 100 DGEBA, 30 DDS, and 25 phenoxy: (a) unetched; (b) etched in chloroform for 15 h

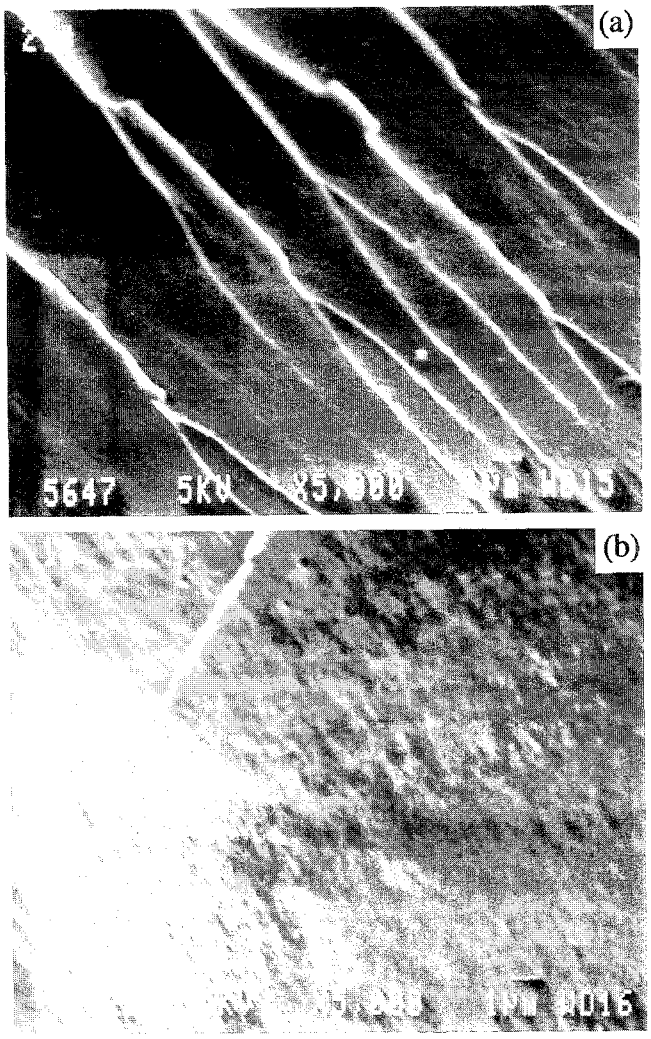


**Figure 8** Fracture surfaces of the cured blend containing 100 DGEBA, 50 MA, and 25 phenoxy: (a) unetched; (b) etched in chloroform for 15 h

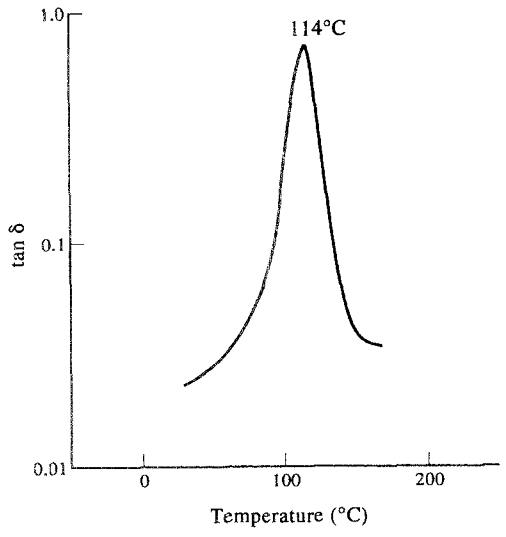


**Figure 7** Dynamic mechanical spectrum of the cured blend containing 100 DGEBA, 50 MA, and 25 phenoxy

results indicate that the DDS-cured ER/phenoxy blend is heterogeneous and that phenoxy is immiscible with DDS-cured ER.

*MA-cured blends*

The cured blend containing 100 DGEBA, 50 MA and 25 phenoxy was transparent; *Figure 7* shows its dynamic mechanical spectrum. This figure clearly displays a maximum (114°C) on the tan $\delta$ vs. $T$ curve, which corresponds to the $T_g$ of the MA-cured blend. The $T_g$ of the latter (114°C) is intermediate between that of the pure phenoxy (91°C) and the neat MA-cured ER (175°C). However, this $T_g$ value for the blend is much lower than that expected for a miscible blend, implying that the cure reaction is incomplete. Scanning electron micrographs of the fractured surface of the MA-cured ER/phenoxy blend (*Figure 8a*) clearly displays some features of brittle facture, but there is no evidence that the blend is heterogeneous. *Figure 8b* shows a micrograph of a specimen that had been etched by chloroform, with no evidence of heterogeneity being observed. It can be concluded that the MA-cured ER/phenoxy blend is homogeneous and that phenoxy is miscible with MA-cured ER.

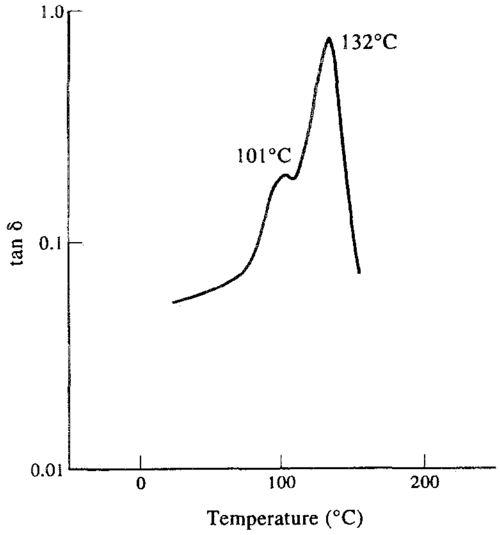


**Figure 9** Dynamic mechanical spectrum of the cured blend containing 100 DGEBA, 75 PA, and 25 phenoxy

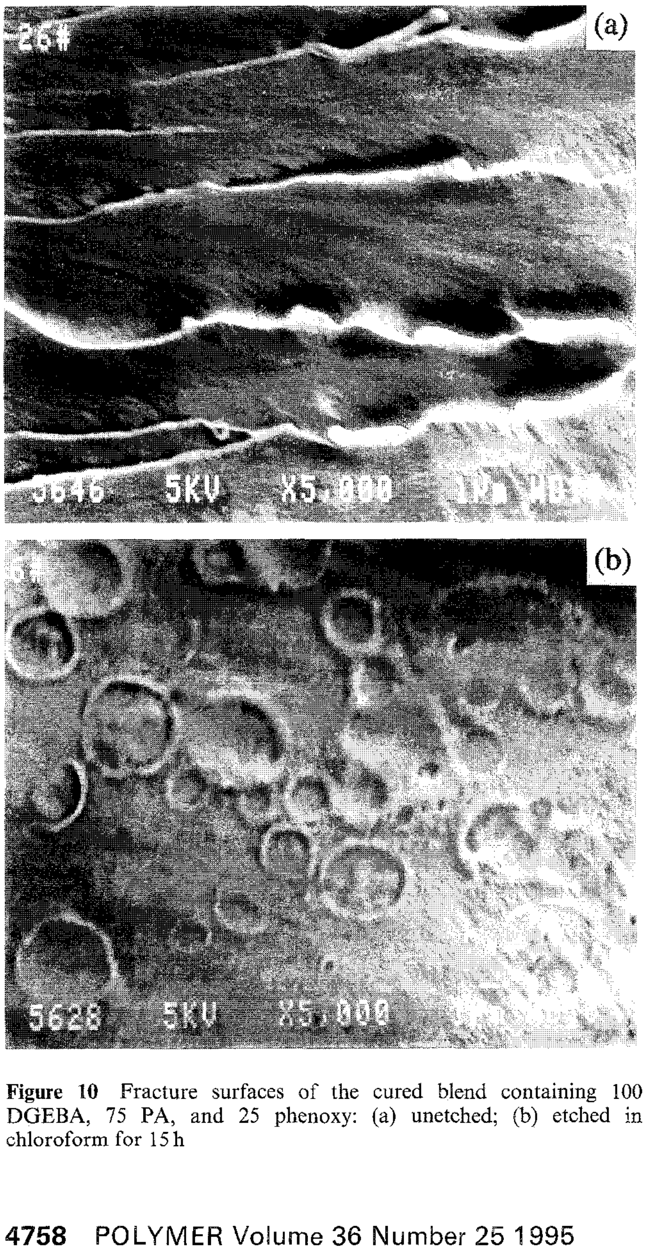


**Figure 10** Fracture surfaces of the cured blend containing 100 DGEBA, 75 PA, and 25 phenoxy: (a) unetched; (b) etched in chloroform for 15 h

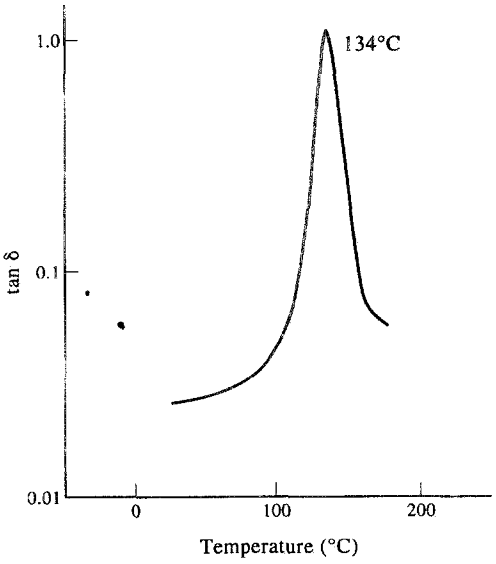


**Figure 11** Dynamic mechanical spectrum of the cured blend containing 100 DGEBA, 80 HHPA, and 25 phenoxy

### *PA-cured blends*

The cured blend containing 100 DGEBA, 75 PA and 25 phenoxy was transparent, indicating that the blend seems to be homogeneous. However, a d.m.a. study clearly revealed two $T_g$s, which are attributed to the phenoxy-rich phase (101°C) and the PA-cured ER phase (132°C), respectively (*Figure 9*). It is obvious that phase separation had occurred in the cured blend and that phenoxy is immiscible with PA-cured ER. It is noted that the $T_g$ value of the PA-cured ER phase (132°C) is very close to that of the neat PA-cured ER (133°C). However, the $T_g$ value of the phenoxy-rich phase (101°C) is higher than that of pure phenoxy (91°C), suggesting that there is some cured ER in the phenoxy-rich phase.

A scanning electron micrograph of the fractured surface of the PA-cured ER/phenoxy blend is given in *Figure 10a*; it appears to show no evidence that the blend is heterogeneous. However, the micrograph of the etched specimen shown in *Figure 10b* clearly reveals a two-phase structure, with domains of the phenoxy-rich phase being dispersed in the continuous phase of the PA-cured, ER-rich phase. The size of the domains is in the range 0.2–5 $\mu$m in diameter. It can be seen that the interface between the phenoxy-rich phase and the PA-cured ER is strongly bonded. It can therefore be concluded that phase separation occurred in the PA-cured ER/phenoxy blend and that phenoxy is not miscible with PA-cured ER.

### *HHPA-cured blends*

The cured blend containing 100 DGEBA, 80 HHPA and 25 phenoxy was transparent, thus indicating the homogeneity of the blend. *Figure 11* shows the dynamic mechanical spectrum of the blend. It can be seen from this figure that only one glass transition peak is observed on the tan $\delta$ vs. $T$ curve, and its value (134°C) is intermediate between that of the pure phenoxy (91°C) and the neat HPPA-cured ER (144°C). These results obviously show that the blend is homogeneous and that phenoxy is miscible with HHPA-cured ER.

The scanning electron micrograph of the fractured

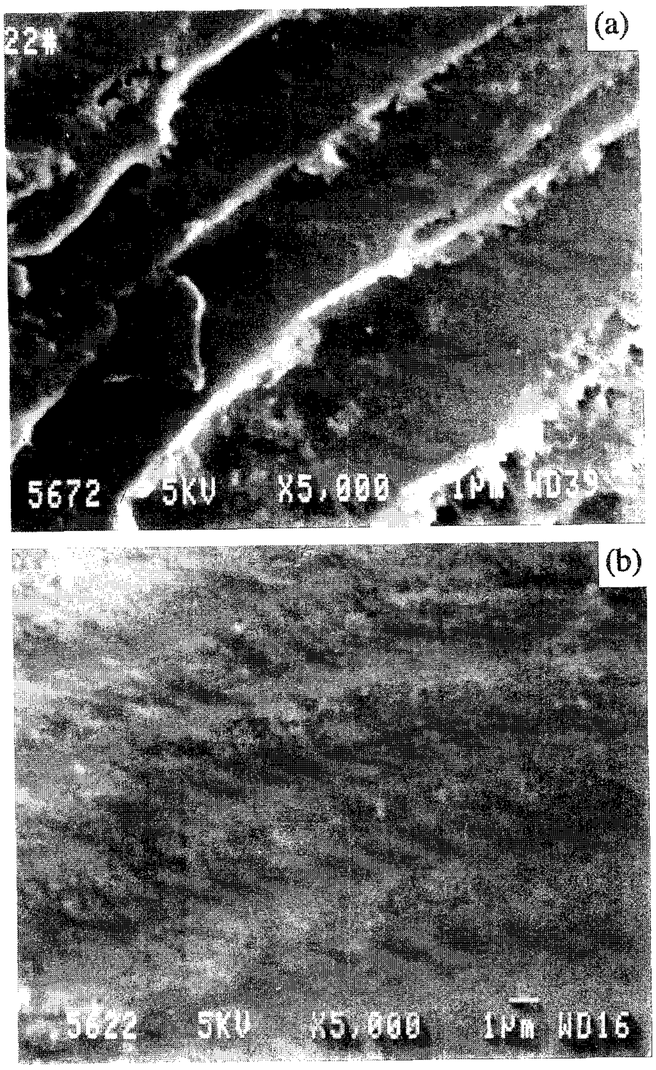


**Figure 12** Fracture surfaces of the cured blend containing 100 DGEBA, 80 HHPA, and 25 phenoxy: (a) unetched; (b) etched in chloroform for 15 h

surface of the HHPA-cured blend (*Figure 12a*) shows no evidence that the blend is heterogeneous; the micrograph of the etched specimen given in *Figure 12b* also displays no characteristic features of an inhomogeneous blend. These observations further confirm that the HHPA-cured ER/phenoxy blend is homogeneous, and that phenoxy is completely miscible with HHPA-cured ER.

## DISCUSSION

The results presented here have shown that phenoxy is miscible over the entire composition range with the uncured ER, i.e. DGEBA, and that the phenoxy/DGEBA blends do not exhibit phase separation up to ~ 280°C. However, phenoxy exhibits different miscibilities with the five cured epoxy resins. The overall compatibility and the resulting morphology of the cured blends are dependent on the choice of cure agent. The cured blends exhibit two categories of phase behaviour: (a) for the blends cured with DDM and aliphatic anhydrides, i.e. MA and HHPA, no phase separation occurs, as indicated by either d.m.s. or SEM; (b) for the blends cured with DDS and aromatic anhydrides, e.g. PA, both techniques clearly show evidence of phase separation.

The formation of a cured ER network caused phase separation in the blends; this can be considered as due to the dramatic change in the chemical and physical nature of the ER during the crosslinking[28,29]. Phase separation is controlled by both thermodynamic and kinetic factors[13].

At first, the effect of crosslinking and the choice of cure agent on the miscibility of the cured blends can be discussed from the viewpoint of thermodynamics. As the cure proceeds, the resulting increase in molecular weight will cause a decrease in the configurational entropy of mixing, so that the enthalpy term, which is usually positive, becomes increasingly more important in determining the free energy of mixing[46], At the same time, changes in the chemical structure which take place during the cure reaction remarkably alter the enthalpy of mixing.

It is interesting to compare the results presented here for the cured ER/phenoxy blends with those previously obtained for the cured ER/PCL blends[36,37] and the cured EP/PES-C blends[35]. It has been proved that both PCL and PES-C are more miscible with the amine-cured epoxy resins than with the anhydride-cured resins. This is because both PCL and PES-C are proton-accepting polymers which offer excellent potential for hydrogen bonding with the hydroxyl groups of the amine-cured epoxy resins which are formed as a result of the opening of the epoxy rings. However, chemical changes in the anhydride-cured ER, such as opening of the epoxy rings, result in the formation of ester groups rather than hydroxyl groups. As a result, the anhydride-cured ER can interact only with proton-donating polymers. Indeed, it is noted that phenoxy exhibits more miscibility with anhydride-cured ER than do either PCL or PES-C[35,37]. The enhanced miscibility is attributed to the presence of pendant hydroxyl groups in the phenoxy, which offer an excellent potential for hydrogen-bonding interactions with the ester groups in the anhydride-cured ER. At the same time, the pendant hydroxyl groups of phenoxy are presumably involved in the cure reaction with anhydride, resulting in interchain crosslinking and the formation of copolymers which can act as compatibilizers for the anhydride-cured ER/phenoxy blends. Nevertheless, although the blends cured with aliphatic anhydrides, i.e. MA and HHPA, are all miscible, the PA-cured ER is immiscible with phenoxy.

For the amine-cured ER/phenoxy blends there is no favorable intermolecular interaction between the components. As a result, phenoxy exhibits less miscibility with the amine-cured epoxy resins than PES-C[35]. Although phenoxy is completely miscible with DDM-cured ER, it becomes immiscible with DDS-cured ER.

On the other hand, differences in the kinetics must also be considered; the choice of hardener will determine the rate of reaction at any given temperature, and hence the period during which phase separation can take place. The rates of diffusion decrease during the curing reaction, and therefore depend directly on the cure kinetics. Differences in reaction kinetics will influence the process of phase separation and thus the resulting morphology of an immiscible cured blend.

## ACKNOWLEDGEMENTS

Financial support from the Presidential Foundation of the Chinese Academy of Sciences and from the State

Science and Technology Commission of China are gratefully acknowledged. This work was also partially sponsored by the University of Science and Technology of China, Hefei.

## REFERENCES

1 Potter, W. G. 'Epoxide Resins', Springer, New York, 1970

2 May, C. A. and Tanaka, Y. (Eds) 'Epoxy Resin Chemistry and Technology', Marcel Dekker, New York, 1973

3 Bauser, R. S. (Ed.) 'Epoxy Resin Chemistry' ACS Symposium Series No. 114, American Chemical Society, Washington DC, 1979

4 Bauser, R. S. (Ed.) 'Epoxy Resin Chemistry II' ACS Symposium Series No. 201, American Chemical Society, Washington DC, 1983

5 Rowe, E. H., Siebert, A. R. and Drake, R. S. *Mod. Plast.* 1970. **47**, 110

6 Sultan, J. M. and MaGarry, F. *Polym. Eng. Sci.* 1973, **13**, 29

7 Drake, R. S., Egan, D. R. and Murphy, W. T. in 'Epoxy Resin Chemistry II' (Ed. R. S. Bauer), ACS Symposium Series No. 221, American Chemical Society, Washington DC, 1982, p. 1

8 Yorkitis, E. M. *et al.* in 'Rubber-Modified Thermoset Resins' (Eds K. Riew and J. K. Gillham), Advances in Chemistry Series No. 208, American Chemical Society, Washington DC, 1984, p. 137

9 Riffle, J. S., Yilgor, I., Banthia, A. K., Tran, C., Wilkes, G. L. and McGrath, J. E. in 'Epoxy Resin Chemistry' (Ed. R. S. Bauer), ACS Symposium Series No. 201, American Chemical Society, Washington DC, 1983, p. 21

10 Cecere, J. A., Hedrick, J. L. and McGrath, J. E. *Am. Chem. Soc. Div. Polym. Chem. Polym. Prepr.* 1985, **26**, 298

11 Kinloch, A. J. in 'Polymer Blends and Mixtures' (Eds D. J. Walsh, J. S. Higgins and A. Maconnochie), Martinus Nijhoff, Dordrecht, 1985, p. 393

12 Diamant, J. and Moulton, R. J. *SAMPE Q.* October 1984, 13

13 Bucknall, C. B. and Partridge, I. K. *Polymer* 1983, **24**, 639

14 Raghava, R. S. *J. Polym. Sci., Polym. Phys. Edn* 1987, **25**, 1017

15 Hedrick, J. L., Hedrick, J. C., Yilgor, I., Wilkes, G. L. and McGrath, J. E. *Adv. Technol. Mater. Process.* 1985, **30**, 947

16 Cecere, J. A. and McGrath, J. E. *Am. Chem. Soc. Div. Polym. Chem. Polym. Prepr.* 1986, **27**, 299

17 Bucknall, C. B. and Gilbert, A. H. *Polymer* 1989, **30**, 213

18 Fu, Z. and Sun, Y. *Chin. J. Polym. Sci.* 1989, **7**, 367

19 Manzione, L. T., Gillham, J. K. and McPherson, C. A. *J. Appl. Polym. Sci.* 1981, **26**, 889

20 Enns, J. B. and Gillham, J. K. *J. Appl. Polym. Sci.* 1983, **28**, 2567

21 Bucknall, C. B., Davies, P. and Partridge, I. K. *Polymer* 1985, **26**, 109

22 Bucknall, C. B. and Partridge, I. K. *Polym. Eng. Sci.* 1986, **26**, 54

23 Verchere, D., Sautereau, H., Pascault, J. P., Moschiar, S. M., Riccardi, C. C. and Williams, R. J. J. *Polymer* 1989, **30**, 107

24 Verchere, D., Sautereau, H., Pascault, J. P., Moschiar, S. M., Riccardi, C. C. and Williams, R. J. J. *J. Appl. Polym. Sci.* 1990, **41**, 467

25 Verchere, D., Pascault, J. P., Sautereau, H., Moschiar, S. M., Riccardi, C. C. and Williams, R. J. J. *J. Appl. Polym. Sci* 1991, **42**, 701

26 Moschiar, S. M., Riccardi, C. C., Williams, R. J. J., Verchere, D., Sautereau, H. and Pascault, J. P. *J. Appl. Polym. Sci.* 1991, **42**, 717

27 Grillet, A. C., Galy, J. and Pascault, J. P. *Polymer* 1992, **34**, 33

28 Guo, Q., Peng, X. and Wang, Z. *Polym. Bull.* 1989, **21**, 593

29 Guo, Q., Peng, X. and Wang, Z. *Polymer* 1991, **32**, 53

30 Guo, Q., Huang, J., Li, B., Chen, T., Zhang, H. and Feng, Z. *Polymer* 1991, **32**, 58

31 Guo, Q., Huang, J., Ge, L. and Feng, Z. *Eur. Polym. J.* 1992, **28**, 405

32 Robeson, L. M., Hale, W. F. and Merriam, C. N. *Macromolecules* 1981, **14**, 1644

33 Coleman, M. M. and Moskala, E. J. *Polymer* 1983, **24**, 251

34 Guo, Q., Huang, J. and Chen, T. *Polym. Bull.* 1988, **20**, 517

35 Guo, Q. *Polymer* 1993, **34**, 70

36 Noshay, A. and Robeson, L. M. *J. Polym. Sci., Polym. Chem. Edn.* 1974, **12**, 689

37 Clark, J. N., Daly, J. H. and Garton, A. *J. Appl. Polym. Sci.* 1984, **9**, 3381

38 Harris, J. E., Goh, S. H., Paul, D. R. and Barlow, J. W. *J. Appl. Polym. Sci.* 1982, **27**, 839

39 Robeson, L. M. and Furtek, A. B. *J. Appl. Polym. Sci.* 1979, **23**, 645

40 Equiazabal, J. I., Iruin, J. J., Carptazar, M. and Guzman, G. M. *Makromol. Chem.* 1984, **185**, 1761

41 Moskala, E. J. and Coleman, M. M. *Polym. Commun.* 1983, **24**, 206

42 Gordon, M. and Taylor, J. S. *J. Appl. Chem.* 1952, **2**, 495

43 Belorgey, G. and Prud'homme, R. E. *J. Polym. Sci., Polym. Phys. Edn.* 1982, **20**, 191

44 Belorgey, G., Aubin, M. and Prud'homme, R. E. *Polymer* 1985, **23**, 1051

45 Grillet, A. C., Galy, J., Pascault, J. P. and Bardin, I. *Polymer* 1989, **30**, 2094

46 Flory, P. J. 'Principles of Polymer Chemistry', Cornell University Press, Ithaca, NY, 1953