PII: S0032-3861(97)00104-3

Polymer Vol. 38 No. 22, pp. 5565–5571, 1997
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0032-3861/97/$17.00+0.00

Miscibility and mechanical properties of epoxy resin/polysulfone blends

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(Received 21 August 1996; revised 17 December 1996)

Bisphenol A-based polysulfone (PSF) was found to be miscible with uncured bisphenol A-type epoxy resin, i.e. diglycidyl ether of bisphenol A (DGEBA), as shown by the existence of a single glass transition temperature (T_g) within the whole composition range. Miscibility between PSF and DGEBA is considered to be due mainly to entropy contribution. Furthermore, PSF was judged to be miscible with the 4,4′-diaminodiphenylmethane (DDM)-cured epoxy resin (ER) as revealed by the means of differential scanning calorimetry (d.s.c.), dynamic mechanical analysis (d.m.a.) and scanning electron microscopy (SEM). D.s.c. and d.m.a. studies showed that the DDM-cured ER/PSF blends had only one T_g. SEM observation revealed that the DDM-cured ER/PSF blends was homogeneous. Both tensile and flexural properties of the DDM-cured ER/PSF blends slightly improved compared to those of the pure DDM-cured ER. Both fracture toughness (K_{IC}) and fracture energy (G_{IC}) increased by *ca.* 20% with the addition of PSF to the system. Morphological investigation of the K_{IC} fracture surface suggests typical characteristics of brittle fracture. © 1997 Elsevier Science Ltd.

(Keywords: epoxy resin; polysulfone; polymer blends)

INTRODUCTION

Epoxy resins are some of the important polymeric materials which are extensively used as the matrices for fibre-reinforced composite materials, adhesives and coatings as well as other engineering materials[1,2]. However, not all the properties of the materials are beneficial, such as inherent brittleness, i.e. low toughness and crack resistance due to their high crosslinking densities. Considerable attention has been paid to modify epoxy resins. A successful modification routine is elastomer toughening, e.g. carboxyl-terminated butadiene-acrylonitrile rubber (CTBN), amine-terminated butadiene-acrylonitrile rubber (ATBN)[3–7], silicone, etc.[8–13] have been applied to enhance toughness of epoxy resins. As a dispersed phase, the elastomeric phase actually acts to alleviate crack propagation and improve the toughness of epoxy resins. In these systems, the toughening has been considered to mainly arise from shear-deformation in the matrices and the presence of rubber particles[9,14–16].

More recently, many attempts have been made to modify epoxy resins with high-performance engineering thermoplastics[16–30], especially to improve toughness of highly crosslinked epoxy resins, e.g. tetrafunctional epoxy resins. Various types of thermoplastics, such as poly(ether sulfone) (PES)[14,17,18], poly(etherimide) (PEI)[16,19–21], poly(ether ether ketone) (PEEK), etc.[22–24], have been explored to modify epoxy resins. Some results indicate that the modifications necessitate a fine phase-separated structure and a good interfacial adhesion between the two separated phases. A thermoplastics-dispersed phase structure or a co-continuous phase structure in thermoplastics-modified epoxy resins usually yields greater fracture toughness. The enhanced toughness of the system has been suggested to arise from crack pinning and the rupture of the dispersed thermoplastics[14,15].

As a part of a series of our studies on the modifications of epoxy resins in this laboratory[23,31–37], this work concerns the investigation on the miscibility and mechanical properties in blends of bisphenol A-type epoxy resin and bisphenol A-based polysulfone (PSF). PSF was chosen as a modifier because of its high toughness, high modulus, high T_g, thermal and chemical resistance and its stability against moisture. Some authors[28–30,38,39] had reported about 4,4′-diaminodiphenylsulfone (DDS)-cured epoxy resin chemically modified with amine and hydroxy terminated polysulfone (PSF) oligomers. They reported that the ductile deformation (stretching and tearing) of the PSF oligomer particles and the plastic deformation of the epoxy matrix around these particles were the main energy absorbing processes. In our work, PSF was physically blended with the epoxy resin (ER) and 4,4′-diaminodiphenylmethane (DDM) was used as the curing agent. The miscibility of both the uncured ER/PSF blends and the DDM-cured ER/PSF blends is examined.

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Furthermore, tensile properties, flexural properties and fracture toughness of the DDM-cured blends are presented.

EXPERIMENTAL

Materials and preparation of samples

The bisphenol A-type epoxy resin E-51 with epoxide equivalent weight 185–210, i.e. diglycidyl either of bisphenol A (DGEBA), was obtained from Wuxi Resin Factory, Wuxi, P.R. China. Bisphenol A-based polysulfone (PSF) was supplied by Shanghai Shuguang Chemical Industrial Factory, Shanghai, P.R. China, it had an intrinsic viscosity $[\eta] = 0.53\,\mathrm{dl\,g^{-1}}$ in the solution of chloroform at 25°C. The PSF was dried at 150°C for 2 h before using. The curing agent was chemically pure grade 4,4′-diaminodiphenylmethane (DDM) and was purchased from Shanghai Reagent Co., Inc., Shanghai, P.R. China.

Uncured DGEBA/PSF blends were prepared by solution casting from chloroform, the solvent was evaporated slowly at room temperature. The residual solvent was removed under vacuum at ambient temperature for 2 weeks.

To prepare the DDM-cured ER/PSF blends, PSF was first dissolved in DGEBA with continuous stirring at 160°C. The mixture obtained was degassed under vacuum for half an hour and then cooled to 100°C. Then, DDM was added to the mixture as the curing agent at 100°C with continuous stirring until a homogeneous ternary mixture was obtained. The ternary mixture was cured in a Teflon mould which was highly polished. The samples were cured successively at 80°C for 2 h, 150°C for 2 h, and 200°C for 2 h.

Differential scanning calorimetry (d.s.c.)

Calorimetric measurement was performed on a Perkin Elmer DSC-7 thermal analysis apparatus in a dry nitrogen atmosphere. The instrument was calibrated with an indium standard. To remove the thermal history of the samples, a thermal pretreatment was used. All samples (about 10 mg in weight) were first heated up to 220°C and held for 3 min, and then quenched to −60°C. All d.s.c. curves were recorded at a heating rate of $20°\mathrm{C\,min^{-1}}$. The glass transition temperatures (T_gs) were taken as the midpoint of the capacity change.

Dynamic mechanical analysis (d.m.a.)

Dynamic mechanical measurements were made on a Dynamic Mechanical Thermal Analyser MK III (Rheomethric Scientific, Ltd., UK) with temperature scanned from 120°C to 200°C. The frequency used was 3 Hz and heating rate $3.0°\mathrm{C\,min^{-1}}$. Specimen dimensions were $1.2 \times 0.2 \times 0.1\,\mathrm{cm^3}$.

Morphological observation

To investigate the phase morphology of the cured ER/PSF blends, the specimens were fractured under cryogenic condition using liquid nitrogen. The fractured surfaces thus obtained were etched in dichloromethane at room temperature for 10 h. The etched specimens were dried to remove the solvent. The K_{IC} fracture surface was also observed. A Hitachi X-650 scanning electron microscope (SEM) was used for observation, before which the surfaces were coated with thin layers of gold of 200 Å.

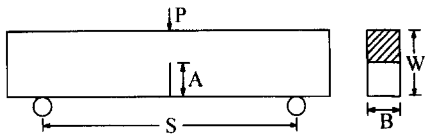

Figure 1 The schematic diagram of three-point bending specimen for measuring fracture toughness

Tensile tests

Tensile tests were carried out on a DCS-5000 testing machine (Shimadzu Co. Ltd., Japan) at ambient temperature (25°C). Standard dumbbell specimens (ASTM D638) with $2.5 \times 0.6 \times 0.4\,\mathrm{cm^3}$ neck were used. A minimum of five specimens were tested in each case to obtain the average value. Crosshead speed was $2\,\mathrm{mm\,min^{-1}}$, corresponding to a relative strain rate of $0.08\,\mathrm{min^{-1}}$.

Flexural tests

The flexural properties were measured by a three-point bending test, according to ASTM D790-84a, on a DCS-5000 testing machine (Shimadzu Co. Ltd., Japan) at ambient temperature (25°C). The specimens were placed on two supports with a span of 64 mm and the load was applied by means of a loading nose midway between the supports. Crosshead speed was $2\,\mathrm{mm\,min^{-1}}$ and a minimum of five specimens were tested in all cases.

Fracture toughness measurements

Fracture toughness was measured by the notched three-point bending test with a crosshead speed of $1.3\,\mathrm{mm\,s^{-1}}$ according to ASTM E399. The schematic diagram of a three-point bending specimen is shown in *Figure 1*. Central Vee-notches were machined in the bars, and extended by pressing a fresh razor blade into the tip of the notch to give a crack length of 3.3 ± 0.3 mm. The thickness of the specimen was about 8 mm and a minimum of five specimens were tested in all cases.

The critical stress intensity factors K_{IC}, i.e. fracture toughness, were calculated using the equation

$$K_{IC} = P_c S / B W^{3/2} f(A/W) \qquad (1)$$

where P_c is the load at crack initiation, B is the thickness of specimen, S is the span width, W is width of the specimen, and A is crack length (see *Figure 1*). The critical strain energy release rate G_{IC}, i.e. fracture energy, was calculated from K_{IC} as

$$G_{IC} = (1 - \gamma^2) K_{IC}^2 / E \qquad (2)$$

where γ is Poisson's ratio and E is the flexural modulus.

RESULTS AND DISCUSSION

Miscibility of DGEBA/PSF blends

All the DGEBA/PSF blends prepared as described above were transparent at ambient temperature. Further heated up to 280°C, these blends still remained clear and no evidence of phase separation was found. This observation suggests that the DGEBA/PSF blends possess a single, homogeneous, amorphous phase.

All the blends were subjected to the studies of thermal analysis. The d.s.c. curves shown in *Figure 2* indicate that all the blends display a single glass transition temperature (T_g), intermediate between those of the two pure components and changing with the blend composition.

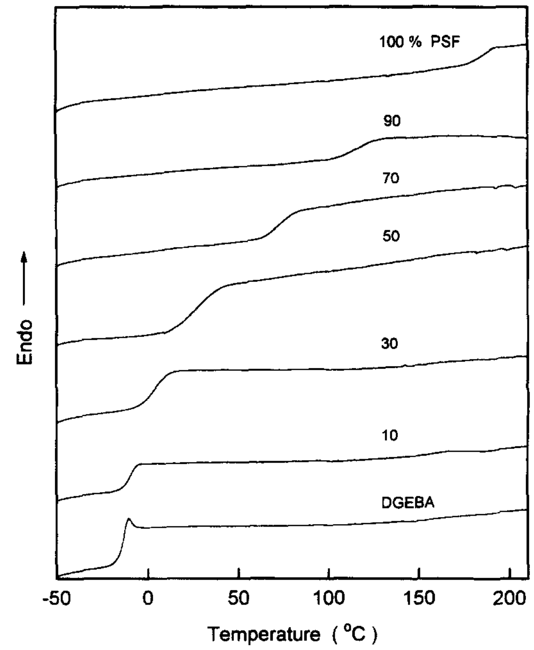

Figure 2 D.s.c. curves of the DGEBA/PSF blends

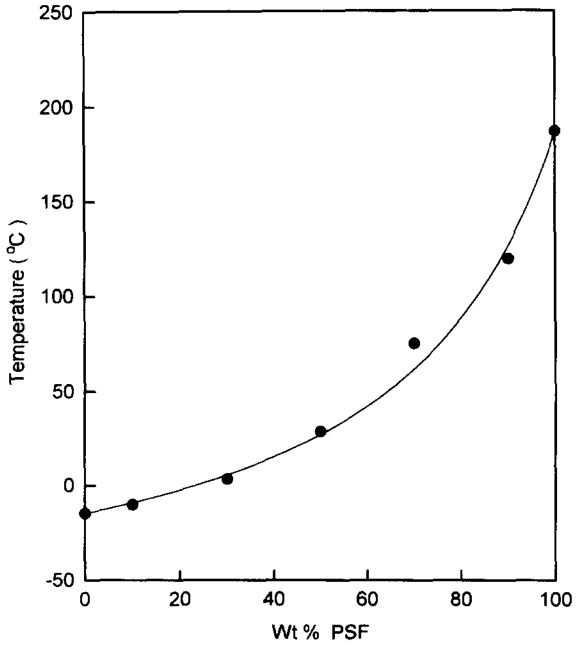

Figure 3 Composition dependence of glass transition temperature of DGEBA/PSF blends. The curve is as predicted by the Gordon–Taylor equation using a k value of 0.26

According to the transparency and glass transition behaviour, the conclusion should be come to that DGEBA/PSF blends are clearly miscible at the entire composition.

Figure 3 summarizes the T_g values obtained from d.s.c. as a function of blend composition. Several theoretical and empirical equations have been used to describe the T_g–composition of miscible polymer blends. One of these, the Gordon–Taylor equation[40], is written as

$$T_g = (W_1 T_{g_1} + kW_2 T_{g_2})/(W_1 + kW_2) \qquad (3)$$

where T_g is the glass transition temperature of the blend, T_{g_1} and T_{g_2} are the glass transition temperatures of components 1 and 2, respectively, W is the weight fraction and k is a constant. The curve in *Figure 3* is drawn using the Gordon–Taylor equation with a k value of 0.26, fitting the experiment data well.

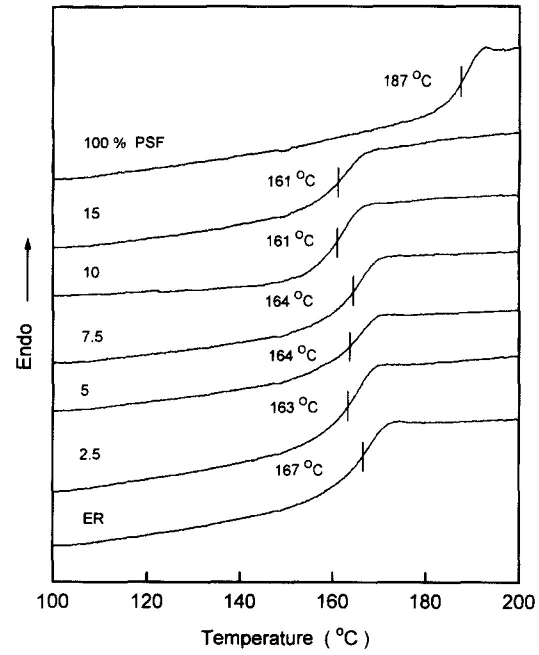

Figure 4 D.s.c. curves of the DDM-cured ER/PSF blends

Prud'homme *et al.*[41,42] have suggested that k can be taken as a semi-quantitative measure of strength of the 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/PSF blends, a k value of 0.26 suggests a weak interaction between DGEBA and PSF. It should be pointed out that the miscibility of the DGEBA/PSF blends can be considered to be due mainly to the entropy contribution as the molecular weight of DGEBA is rather low.

Miscibility of DDM-cured ER/PSF blends

All the DDM-cured ER/PSF blends studied were transparent at room temperature. Further heated up to 300°C, these blends still remained clear. This observation primarily suggests that these blends were homogeneous and had single-phase structure.

D.s.c. The d.s.c. curves of the DDM-cured ER/PSF blends all give one single T_g, as shown in *Figure 4*. In addition, all the blends studied remained essentially transparent even at 300°C. The existence of a single T_g, together with the transparency, suggests the homogeneous nature of the DDM-cured ER/PSF blends.

From *Figure 4*, it can be seen that the T_g of the blends are all somewhat lower than that of the pure DDM-cured ER. The addition of PSF raised the viscosity of the system, which could result in an incomplete curing reaction due to the steric hindrance under the present curing condition[43]. Furthermore, the dilution effect of PSF is also an important factor which gives rise to the decrease of the crosslinking density. Therefore, the crosslinked network gives lower glass transition temperatures.

Dynamic mechanical properties. Presented in *Figure 5* are the dynamic mechanical spectra ranging from 120 to

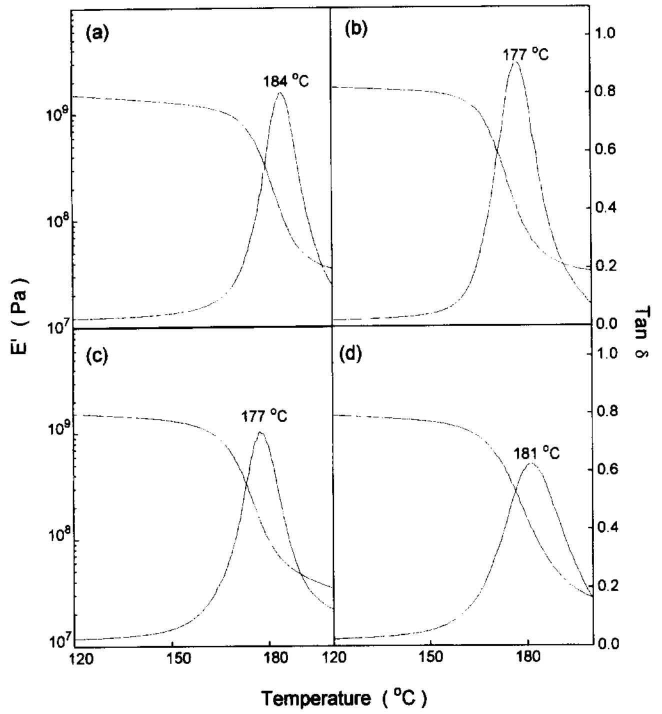

Figure 5 Dynamic mechanical spectra of (a) 100/0, (b) 95/5, (c) 90/10 and (d) 85/15 DDM-cured ER/PSF blends

200°C for the pure DDM-cured ER and the DDM-cured ER/PSF blends. *Figure 5a* shows the dynamic mechanical spectrum of the pure DDM-cured ER, and there exists a well-defined relaxation peak centred at 184°C, which is ascribed to the glass transition of the cured ER. *Figures 5b–d* show the dynamic mechanical spectra of DDM-cured ER/PSF blends containing 5, 10 and 15 wt% PSF, respectively. A single tan δ peak could be identified for these ER/PSF blends in *Figures 5b–d*. This result confirms the full miscibility of the blend components. It can be seen that the T_g of ER/PSF blends obtained by d.m.a. are also slightly lower than that of the pure DDM-cured ER. The d.m.a. result presented here is in good agreement with that from d.s.c.

It can also be seen from *Figure 5* that the relative width of half-height of glass transition peak remarkably increases with increasing PSF content. The width of the glass transition can reflect the magnitude of local composition fluctuations in the polymer blends, and hence the relative homogeneity or miscibility of the system[44]. The peak width increasing with PSF content also implies that the DDM-cured ER/PSF blends are miscible.

Phase morphology. The morphology of the cured blends were investigated by means of SEM. *Figure 6* presents the SEM micrographs of solvent-etched fracture ends of the 92.5/7.5 and 85/15 DDM-cured ER/PSF blends. The specimens were fractured under cryogenic condition using liquid nitrogen and etched with dichloromethane. If the cured blends were phase-separated, the PSF phase could be preferentially etched by the solvent while the cured ER phase remained unaffected. The SEM micrographs in *Figure 6* appear to show no evidence that the blends are heterogeneous. The SEM observation supports the d.s.c. and d.m.a. results.

From the above results, it can reasonably be concluded that all the DDM-cured ER/PSF blends obtained are homogeneous and have a single phase. These results are different to McGrath's[27–29,38]. They found that DDS-cured ER/PSF oligomer blends were immiscible. It has been shown that the different curing agents could have a remarkable influence on the phase behaviour of the epoxy blends[34,35].

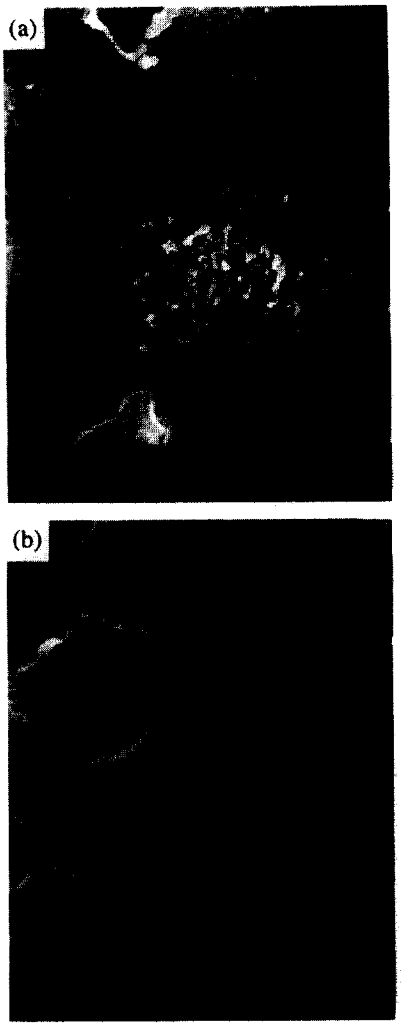

Figure 6 SEM micrographs of fractured surfaces of (a) 92.5/7.5 and (b) 85/15 DDM-cured ER/PSF blends etched with dichloromethane

With DDM adding to DGEBA/PSF mixture at elevated temperature, the curing reaction involving chain extension, branching and crosslinking took place. As the curing reaction proceeded, the molecular weight of the system greatly increased, and the crosslinking network structure in three-dimensions formed. On the other hand, the occurrence of crosslinking caused the dramatic changes of the chemical and physical natures in the system. Both the factors affected the phase behaviour of the blends. Of course, competition exists between the phase separation and the crosslinking reaction during the curing process. If gelation occurs prior to phase separation, homogeneous blends can be obtained. In the present case, the relatively low initial curing temperature (80°C) and the high viscosity of the PSF are beneficial for obtaining homogeneous blends, and therefore a semi-interpenetrating polymer network (semi-IPN) may form with the system remaining miscible. Furthermore, it is the homogeneous phase structure that resulted in the decrease of the crosslinking density and hence the decrease of T_g of the DDM-cured blends.

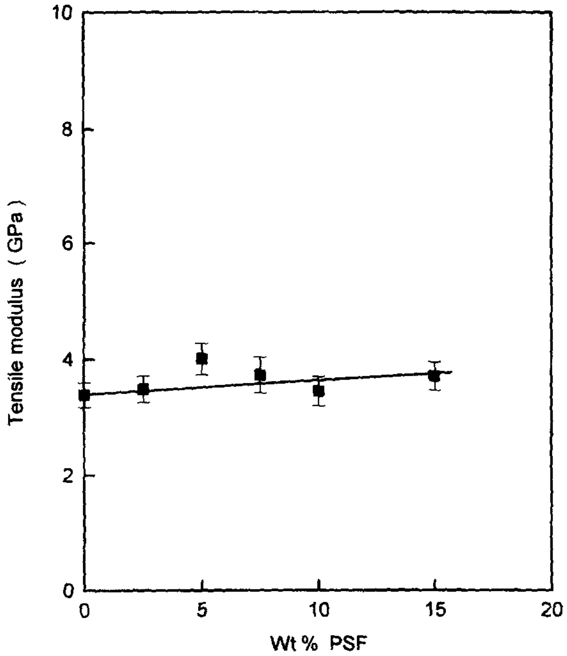

Figure 7 Tensile modulus as a function of PSF content for the DDM-cured ER/PSF blends

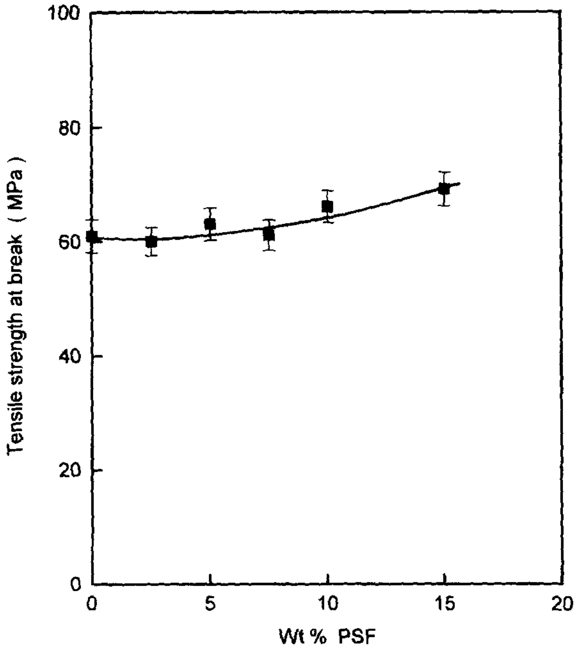

Figure 8 Tensile strength at break as a function of PSF content for the DDM-cured ER/PSF blends

Mechanical properties of DDM-cured ER/PSF blends

Tensile properties. No yields were observed on the stress–strain curves of the pure DDM-cured epoxy resin and all the blends, showing that DDM-cured ER and its blends with PSF were basically brittle materials at room temperature. From the initial slopes of the stress–strain curves, tensile moduli were calculated. The values obtained for tensile modulus, tensile strength at break and elongation at break are plotted in *Figures 7–9*, respectively, as functions of PSF content. It can be seen that tensile modulus, tensile strength at break and elongation at break all show basically a slight increase with increase of PSF content. The addition of PSF slightly improved the tensile properties of the DDM-cured ER.

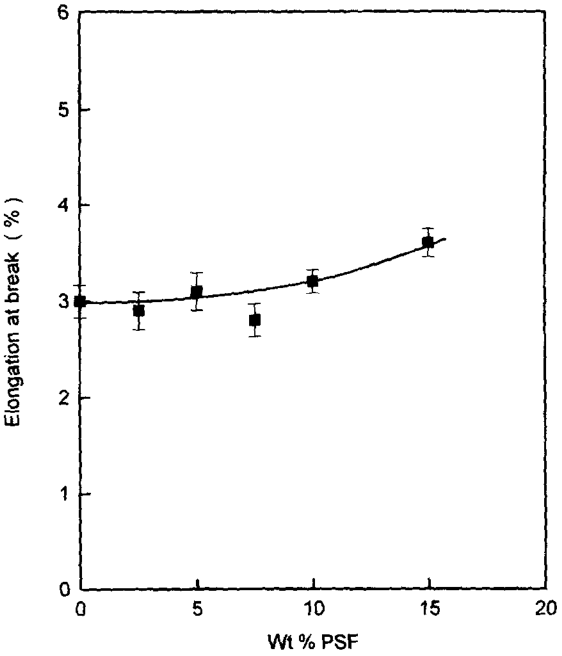

Figure 9 Elongation at break as a function of PSF content for the DDM-cured ER/PSF blends

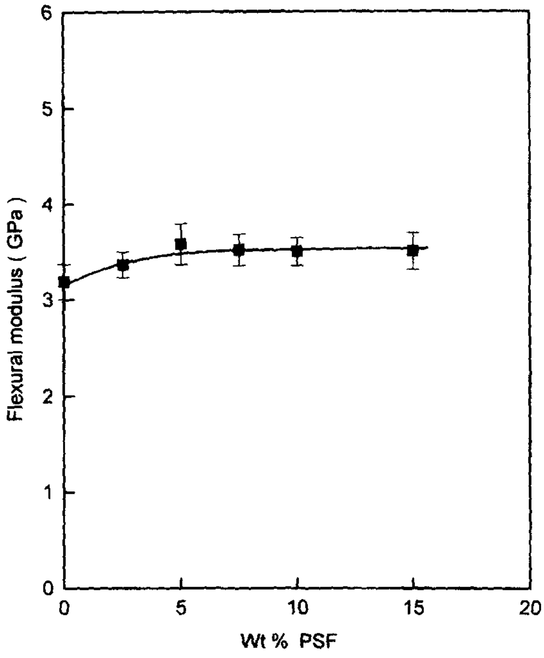

Figure 10 Flexural modulus as a function of PSF content for the DDM-cured ER/PSF blends

Flexural properties. Figures 10–12 respectively show the flexural modulus, flexural strength at break and flexural strain at break as functions of PSF content for the DDM-cured ER/PSF blends. They all show a modest increase with increasing PSF content. McGrath *et al.* had reported that flexural modulus decreased slightly when modified with amine and hydroxy terminated PSF oligomers[27–29,38]. This difference in flexural modulus between ours and McGrath's is because the PSF we used is simply a polymer, but not an oligomer.

Fracture toughness. The plots of K_{IC} and G_{IC} vs. PSF content in the cured blends are presented in *Figure 13*. As observed, both the fracture toughness (K_{IC}) and the fracture energy (G_{IC}) increased by *ca.* 20% with the addition

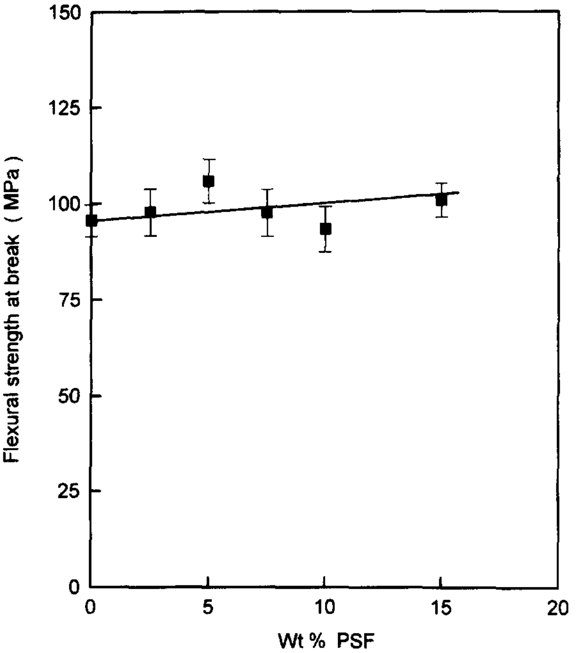

Figure 11 Flexural strength at break as a function of PSF content for the DDM-cured ER/PSF blends

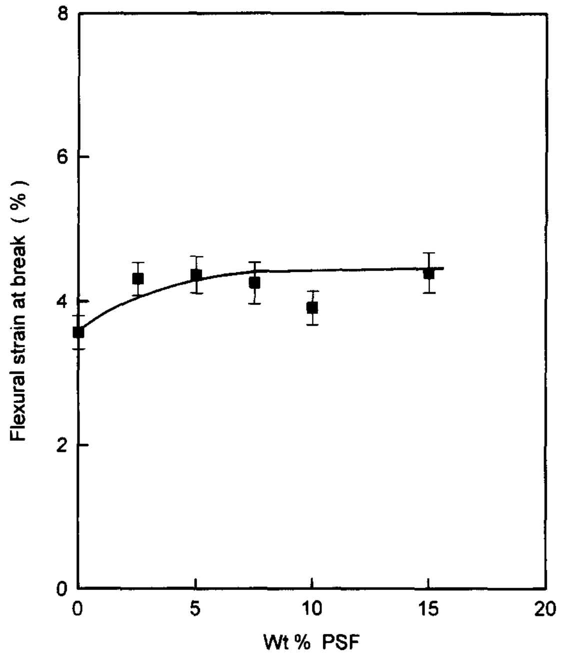

Figure 12 Flexural strain at break as a function of PSF content for the DDM-cured ER/PSF blends

of PSF to the system. We could not assume that the toughening effect was completely due to the contribution of the thermoplastic polymer. A decrease in crosslinking density of the epoxy network could also lead to the same behaviour[45].

The SEM micrograph of the surface of fracture mechanics test end can give some information about the toughening mechanism. Shown in *Figure 14* is an SEM micrograph of the K_{IC} fracture surface of the 85/15 DDM-cured ER/PSF blend. The cracks spread freely and regularly, and oriented in the direction of loading, suggesting typical characteristics of brittle fracture. It appears to be rather different from that observed by McGrath *et al.*[27–29,38]. They used hydroxyl terminated PSF oligomers and obtained the DDS-cured blends with two-phase structure and good interfacial adhesion, showed a ductile deformation of the PSF particles and a plastic deformation of the DDS-cured epoxy matrix around these particles.

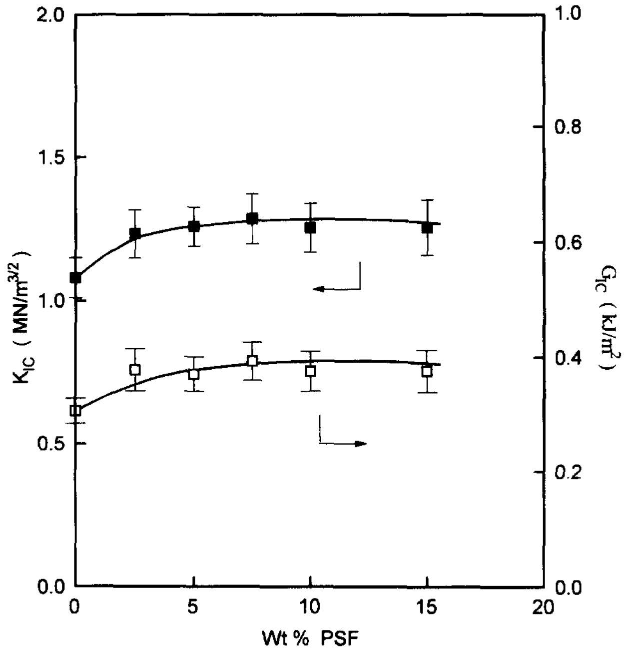

Figure 13 K_{IC} and G_{IC} as functions of PSF content for the DDM-cured ER/PSF blends

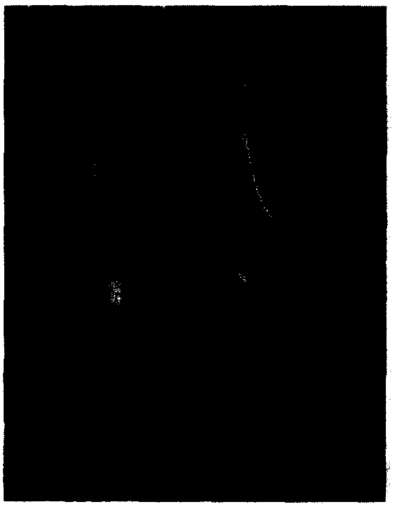

Figure 14 SEM micrograph of K_{IC} fracture surface of the 85/15 DDM-cured ER/PSF blend

CONCLUSIONS

The results presented here clearly show that DGEBA/PSF blends are completely miscible over the entire composition. The miscibility between DGEBA and PSF is mainly attributed to entropy contribution. D.s.c., d.m.a. and SEM studies revealed that the DDM-cured ER/PSF blends obtained were homogeneous. Both tensile and flexural properties of the DDM-cured ER/PSF blends slightly improved compared to those of the pure DDM-cured ER. Both K_{IC} and G_{IC} increased by *ca.* 20% with the addition of PSF to the system. Morphological investigation of the K_{IC} fracture surface of the 85/15 DDM-cured ER/PSF blend shows typical characteristics of brittle fracture.

ACKNOWLEDGEMENTS

The financial support from the Presidential Fund of the Chinese Academy of Sciences and from the State Science and Technology Commission of China. The authors also wish to express their appreciation to the National Natural Science Foundation of China for awarding a 'Premier' Grant for Outstanding Young Scientists (No. 59525307).

REFERENCES

1. May, C. A. and Tanaka, G. Y. (ed.), *Epoxy Resin Chemistry and Technology*. Marcel Dekker, New York, 1973.
2. Bauer, R. S. (ed.), in *Advances in Chemistry*, Vol. 114. American Chemical Society, Washington, DC, 1979.
3. Bucknall, C. B., *Toughened Plastics*. Applied Science, London, 1977.
4. Sultanm, J. N. and McGarry, F. J., *Polym. Eng. Sci.*, 1973, **13**, 29.
5. Kunz, S. C., Sayre, J. A. and Assink, R. A., *Polymer*, 1982, **23**, 1897.
6. Sasidaran, A. P., Latha, P. B. and Ramaswamy, R., *J. Appl. Polym. Sci.*, 1990, **41**, 15.
7. Yee, A. F. and Pearson, R. A., *J. Mater. Sci.*, 1986, **21**, 2462.
8. Yorkgitis, E. M., Eiss, N. S., Tran, C. Jr., Wilkes, G. L. and McGrath, J. E., *Am. Chem. Soc. Adv. Polym. Sci.*, 1985, **72**, 79.
9. Riew, C. K., Rowe, E. H. and Siebert, A. R., *Am. Chem. Soc., Adv. Chem. Ser.*, 1976, **154**, 326.
10. Meijerink, J. I., Eguchi, S., Ogata, M., Ishii, T., Amagi, S., Numata, S. and Sashima, H., *Polymer*, 1994, **35**, 179.
11. Zheng, S., Wang, H., Dai, Q., Luo, X., Ma, D., Wang, K., *Makromol. Chem.*, 1995, **196**, 269.
12. Kemp, T. J., Wilford, A., Howarth, O. W. and Lee, T. C. P., *Polymer*, 1992, **33**, 1860.
13. Bussi, P. and Ishida, H., *J. Appl. Polym. Sci.*, 1994, **32**, 647.
14. McGarry, F. J., *Proc. Roy. Soc., Lond.*, 1970, **A319**, 59.
15. Kinlock, A. J. and Young, R. J., *Fracture Behaviour of Polymers*. Applied Science, New York, 1989.
16. Diamont, J. and Moulton, R. J., *29th National SAMPE Symposium*, California, 1984, p. 422.
17. Raghava, R. S., *J. Polym. Sci., Polym. Phys. Edn.*, 1987, **25**, 1017.
18. Raghava, R. S., *J. Polym. Sci., Polym. Phys. Edn.*, 1988, **26**, 65.
19. Hourston, D. J. and Lane, J. M., *Polymer*, 1992, **33**, 1397.
20. Chen, M. C., Hourston, D. J. and Sun, W. B., *Eur. Polym. J.*, 1995, **31**, 199.
21. Cho, J. B., Hwang, J. W., Cho, K., An, J. H. and Park, C. E., *Polymer*, 1993, **34**, 4832.
22. Iijima, T., Tochiomoto, T. and Tomoi, M., *J. Appl. Polym. Sci.*, 1991, **63**, 311.
23. Guo, Q., Huang, J., Li, B., Chen, T., Zhang, H. and Feng, Z., *Polymer*, 1991, **32**, 58.
24. Yee, A. F. and Pearson, R. A., *Polym. Mater. Sci. Eng.*, 1990, **63**, 311.
25. Bucknall, C. B. and Partridge, I. K., *Polymer*, 1983, **24**, 639.
26. Bucknall, C. B. and Gilbert, A. H., *Polymer*, 1989, **30**, 213.
27. Gilbert, A. H. and Bucknall, C. B., *Makromol. Chem., Macromol. Symp.*, 1991, **45**, 289.
28. Hedrick, J. L., Yilgor, I., Jurek, M., Hedrick, J. C., Wilkens, G. L. and McGrath, J. E., *Polymer*, 1991, **32**, 2020.
29. Cecere, J. A. and McGrath, J. E., *Polym. Prepr.*, 1986, **27**, 299.
30. Hedrick, J. L., Yilgor, I., Wilkens, G. L. and McGrath, J. E., *Polym. Bull.*, 1985, **13**, 201.
31. Guo, Q., Peng, X. and Wang, Z., *Polym. Bull.*, 1989, **21**, 593.
32. Guo, Q., Peng, X. and Wang, Z., *Polymer*, 1991, **32**, 53.
33. Guo, Q., Huang, J., Ge, L. and Feng, Z., *Eur. Polym. J.*, 1992, **28**, 405.
34. Guo, Q., *Polymer*, 1993, **34**, 70.
35. Guo, Q., *Polymer*, 1995, **36**, 4753.
36. Zheng, S., Hu, Y., Guo, Q. and Wei, J., *Colloid Polym. Sci.*, 1996, **274**, 410.
37. Zheng, S., Wang, J., Guo, Q., Wei, J. and Li, J., *Polymer*, 1996, **37**, 4667.
38. Hedrick, J. L., Jurek, M. J., Yilgor, I. and McGrath, J. E., *Polym. Prepr.*, 1985, **26**(2), 293.
39. Fu, Z. and Sun, Y., *Chinese J. Polym. Sci.*, 1989, **7**, 367.
40. Gordon, M. and Taylor, J. S., *J. Appl. Chem.*, 1952, **2**, 495.
41. Belorgey, G. and Prud'homme, R. E., *J. Polym. Sci., Polym. Phys. Edn.*, 1982, **20**, 191.
42. Belorgey, G., Aubin, M. and Prud'homme, R. E., *Polymer*, 1982, **23**, 1051.
43. Zheng, S., Zhang, N., Luo, X. and Ma, D., *Polymer*, 1995, **36**, 3609.
44. MacKnight, W. J., Karasz, F. E. and Fried, J. R., in *Polymer Blends*, Vol. 1, Chapter 5, ed. D. R. Paul and S. Newman. Academic Press, New York, 1978.
45. Pearson, R. A. and Yee, A. E., *Tough Compos. Mater. Rec. Dev.* Noyes, Park Ridge, NJ, 1987, pp. 157–177.