

The Preparation and Morphology of PPO-Epoxy Blends

RAYMOND A. PEARSON*[†] and ALBERT F. YEE

Department of Materials Science and Engineering, University of Michigan, Ann Arbor, Michigan 48109

SYNOPSIS

Poly(phenylene oxide) (PPO) was found to be miscible in diglycidyl ether of bisphenol A (DGEBA) based epoxy. The PPO-DGEBA system exhibited upper critical solution temperature (UCST) behavior. The cloud point temperatures were measured and found to be sensitive to the mol wt of the epoxy resin. A series of PPO-modified epoxies were cured with piperidine at 160°C, which is above the cloud point temperature. Upon cure, two phase solids were formed, which contained discrete PPO particles. However, the two-phase particulate morphology was not uniform and numerous large, occluded PPO particles were observed. In order to improve the uniformity, several styrene-maleic anhydride copolymers were evaluated as potential surfactants for PPO-DGEBA blends. The formation of a uniform, particulate morphology was facilitated by the addition of a styrene-maleic anhydride copolymer, containing a 10 : 1 ratio of styrene to maleic anhydride. To our knowledge, this is the first time that an emulsifying agent has been added to improve the morphology of thermoplastic modified epoxies. © 1993 John Wiley & Sons, Inc.

INTRODUCTION

The technology of toughening epoxies with thermoplastic particles is a relatively new concept. Since the pioneering work of Bucknall and Partridge¹ on epoxies modified with poly(ether sulfone), there has been much activity in this area, as suggested by the number of articles in the scientific literature.¹⁻¹³ Most of the focus of such work addresses the amount of toughness obtained and those material parameters that enhance the toughening effect. It is useful to divide these works into two groups. In the first group, custom-made reactive oligomers were utilized to toughen epoxies, while in the other, commercial thermoplastic resins were used. Both approaches for producing thermoplastic-toughened epoxies have shown moderate success as long as a two phase morphology is observed. Unfortunately, there has been little progress made in determining the toughening mechanisms in these novel materials, and even less progress in controlling the morphology.

The purpose of this work is to prepare a new type of thermoplastic modified epoxy, a poly(phenylene oxide)-epoxy blend. PPO was chosen to modify a ductile DGEBA-piperidine epoxy system, since its refractive index (at room temperature) closely matches that of the DGEBA-piperidine epoxy system. The result is a transparent material, which should lend itself to optical microscopic observations of the toughening mechanism(s). The toughening effect and toughening mechanism(s) will be reported in a subsequent article.¹⁴ For this article, we shall focus on the synthetic means used to produce these epoxies and describe the use of a styrene-maleic anhydride copolymer to stabilize the particulate morphology of these two phase materials.

EXPERIMENTAL

Materials

All materials used throughout this work were from commercial sources and were used as received. The epoxy system consisted of a diglycidyl ether of bisphenol-A based epoxy (DGEBA), cured with piperidine. Two types of DGEBA epoxy were used, which were DER 331 and DER 332 resins, obtained from the Dow Chemical Company. This epoxy sys-

* Present address: Lehigh University, Whitaker Lab #5, Bethlehem, Pennsylvania 18015.

[†] To whom correspondence should be addressed.

tem was modified by dissolving polyphenylene oxide (PPO) in the hot epoxy. Two types of PPO resins were examined, which were PPO-1 and PPO-2 resins from the General Electric Company. PPO-1 resin is a standard injection molding grade material with a mol wt of 42,500 g/mole. PPO-2 resin is a low viscosity injection molding grade with a mol wt of 34,000 g/mole.

The Epoxy-PPO alloys investigated were comprised of two glassy phases, where PPO was the particulate phase. Unfortunately, the particulate morphology was not uniform. Therefore, the work reported here focuses on the stabilization of the particulate morphology. Several parameters controlling the morphology of PPO-modified epoxies were examined: (1) the PPO mol wt, (2) the addition of low mol wt styrene-maleic anhydride block copolymers, (3) the addition of high mol wt styrene-maleic anhydride block copolymers, (4) the mol wt of the DGEBA resin, and (5) PPO content.

A list of materials used in this work can be found in Table I, and the chemical structures are shown in Figure 1.

Chemical Preparation and Apparatus

A schematic diagram of the apparatus used to prepare the blends is shown in Figure 2. The list of equipment includes (1) a disposable flint glass reactor, (2) a lab jack for raising and lowering the reactor, (3) a mechanical stirrer, (4) a heating mantle, (5) a type J thermocouple connected to a

temperature indicator, (6) a vacuum line with vacuum gage, and (7) a vacuum pump. The apparatus allows the PPO/epoxy formulations to be mixed under controlled heating. The thoroughly mixed and degassed resin is poured into a preheated mold and cured for sixteen hours at 160°C.

Series 1: The Effect of PPO Mol Wt

The two PPO resins, supplied by the General Electric Co., having weight average mol wts of 42,500 g/mole and 34,000 g/mole, respectively, were examined. The PPO powder, at a concentration of 10 parts per hundred epoxide resin (phr), was dissolved into the epoxy by mechanically stirring the mixture at 190°C for 2 h. The solution was cooled to 150°C and the curing agent piperidine was added using a syringe. The needle of the syringe was submerged in the hot epoxy to reduce the amount of piperidine that boiled off. The solution was degassed and was poured into a preheated mold and cured for 16 h at 160°C in an air-circulating oven. The mold was removed from the oven and was allowed to cool to room temperature.

Series 2: The Use of SMA Diblock Copolymers

A series of oligomeric styrene-maleic anhydride block copolymers, from the Sartomer Co., was evaluated as possible surfactants. Since polystyrene is miscible in PPO and maleic anhydride reacts with epoxy, SMA block copolymers are expected to be good surfactants between the two phases. Three

Table I List of Materials

Product Code	Designation	Description	Mol Wt	PS/MA Ratio
DER 332 Resin ^a	DGEBA-1	DGEBA-Based Epoxy	342	—
DER 331 Resin ^a	DGEBA-2	DGEBA-Based Epoxy	378	—
DER 337 Resin ^a	DGEBA-3	DGEBA-Based Epoxy	480	—
Piperidine ^b	PIP	Secondary Amine	80	—
PPO-1 Resin ^c	PPO-1	Poly(phenylene oxide)	34,000	—
PPO-2 Resin ^c	PPO-2	Poly(phenylene oxide)	42,500	—
SMA 1000 Resin ^d	SMA-1	SMA Block Copolymer	16,000	1/1
SMA 2000 Resin ^d	SMA-2	SMA Block Copolymer	17,000	2/1
SMA 3000 Resin ^d	SMA-3	SMA Block Copolymer	18,000	3/1
DYLARK 332 Resin ^e	SMA-4	SMA Copolymer	180,000	4.5/1
DYLARK 232 Resin ^e	SMA-5	SMA Copolymer	231,000	7.5/1
DYLARK 132 Resin ^e	SMA-6	SMA Copolymer	260,000	10/1

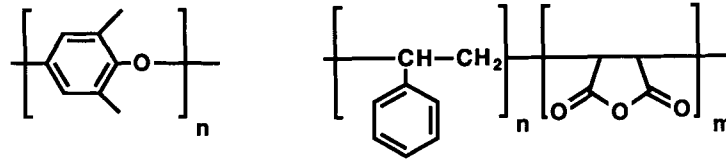
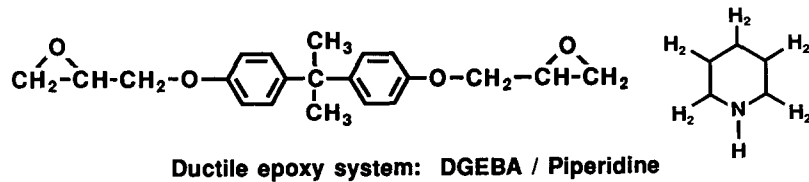
^a Supplied by the Dow Chemical Company.

^b Supplied by the Fischer Chemical Company.

^c Supplied by the General Electric Company.

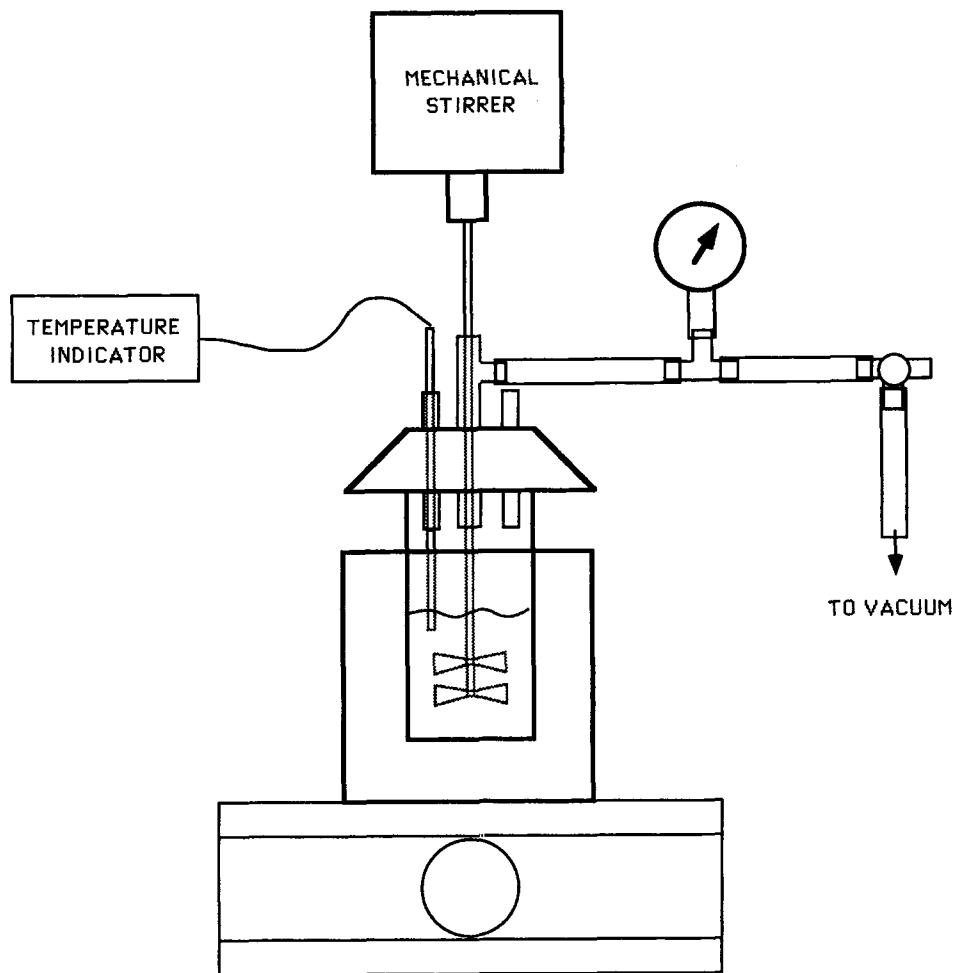
^d Supplied by the Sartomer Chemical Company.

^e Supplied by the ARCO Chemical Company.

**Rigid modifier: PPO****Copolymer: SMA****Figure 1** Chemical structures for the materials used in this investigation.

types of SMA resins were examined: SMA-1 (PS/MA ratio = 1/1; MW = 1600), SMA-2 (PS/MA ratio = 2/1; MW = 1700), and SMA-3 (PS/MA-

3/1; MW = 1800). These SMA resins were added at the level of 1 phr. Higher concentrations of SMA-3 were also examined since it appeared to produce

**Figure 2** Schematic diagram of epoxy preparation apparatus.

the best results. PPO and SMA were added to the epoxy at room temperature and were heated to 200°C while stirring. After the SMA and PPO were dissolved, the solution was then cooled to 150°C. Piperidine was added at 150°C and the remainder of the cure conditions was the same as described above.

Series 3: The Use of SMA Random Copolymers

Higher mol wt SMA random copolymers, from the ARCO Chemical Co., were tried as possible surfactants for PPO-modified epoxies: SMA-4 (PS/MA = 4.5/1), SMA-5 (PS/MA = 7.5/1), and SMA-6 (PS/MA = 10/1). PPO and SMA were added to the epoxy at room temperature and were heated to 200°C while stirring. After the SMA and PPO were dissolved into the epoxy, the solution was then cooled to 150°C. Piperidine was added at 150°C and the remainder of the cure conditions were the same as described above.

The formulations for series 1–3 are shown in Table II.

Series 4: The Effect of DGEBA Mol Wt

Three different DGEBA resins, from the Dow Chemical Co., with average mol wts of 348, 372, and 480 g/mole, respectively, were examined. The curing conditions were the same as those described above.

In addition to preparing plaques for mechanical measurements, the cloud point temperatures of these

DGEBA–PPO mixtures were measured using an optical microscope equipped with a heating stage. The cloud point temperatures of larger samples were determined by visual observation of the reactor during the cooling period.

Series 5: The Effect of PPO Concentration

Four PPO concentrations were examined: 5, 10, 15, and 30 phr. The combination of the lowest mol wt epoxy, the lowest mol wt PPO resin, and the SMA-6 copolymer were used in an effort to stabilize the PPO morphology. PPO and SMA were added to the epoxy at room temperature and were heated to 200°C while stirring. After the SMA and epoxy were dissolved, the mixture was then cooled to 150°C. The remainder of the curing conditions were the same as those described above.

The formulations for series 4–5 are shown in Table III.

Analysis

The morphology of the cured plaques was determined by visual examination, optical microscopy, and electron microscopy. In some cases, visual inspection of the fracture surface was sufficient to assess the morphology of the material, since improperly stabilized formulations exhibited gross phase separation. Differential scanning calorimetry was used to assess any shifts in the glass transition temperature that would be indicative of miscibility.

Table II Formulations for Series 1–2

Designation	DGEBA-2 (g)	Piperidine (mL)	PPO (g)	SMA (g)	CTBN (g)
Series 1: The Effect of PPO Mol Wt					
DGEBA/PIP	500	29	—	—	—
DGEBA/PIP/PPO-1 (10)	500	29	50 ^a	—	—
DGEBA/PIP/PPO-2 (10)	500	29	50 ^b	—	—
Series 2: The Use of Low MW SMA Copolymers					
DGEBA/PIP	500	29	—	—	—
DGEBA/PIP/PPO (10)	500	29	50 ^a	—	—
DGEBA/PIP/PPO (10)/SMA-1 (1)	500	29	50 ^a	5	—
DGEBA/PIP/PPO (10)/SMA-2 (1)	500	29	50 ^a	5	—
DGEBA/PIP/PPO (10)/SMA-3 (1) ^c	500	29	50 ^a	5	5
DGEBA/PIP/PPO (10)/SMA-3 (5) ^c	500	29	50 ^a	25	5
DGEBA/PIP/PPO (10)/SMA-3 (10) ^c	500	—	50 ^a	50	5

^a Denotes that PPO⁻¹ resin was used.

^b Denotes that PPO⁻² resin was used.

^c Denotes that 1 phr CTBN has been added to aid in the visual examination of the morphology.

Table III Formulations for Series 3 and 5

Designation	DGEBA-2 (g)	Piperidine (mL)	PPO (g)	SMA (g)
Series 3: The Use of High Mol Wt SMA Copolymers				
DGEBA/PIP	500	29	—	—
DGEBA/PIP/SMA-4 (5)	500	29	—	25
DGEBA/PIP/SMA-5 (5)	500	29	—	25
DGEBA/PIP/SMA-6 (5)	500	29	—	25
DGEBA/PIP/PPO (10)/SMA-4 (5)	500	29	50 ^a	25
DGEBA/PIP/PPO (10)/SMA-5 (5)	500	29	50 ^a	25
DGEBA/PIP/PPO (10)/SMA-6 (5)	500	29	50 ^a	25
DGEBA/PIP/SMA-5 (P)	500	29	—	??
DGEBA/PIP/PPO (10)/SMA-5 (P) ^b	500	29	50 ^a	??
Series 5: The Effect of PPO Content				
DGEBA/PIP	500	29	—	—
DGEBA/PIP/SMA-5 (2.5)/PPO (5)	500	29	25 ^a	12.5
DGEBA/PIP/SMA-6 (2.5)/PPO (10)	500	29	50 ^a	12.5
DGEBA/PIP/SMA-6 (2.5)/PPO (15)	500	29	75 ^a	12.5
DGEBA/PIP/SMA-6 (2.5)/PPO (~20) ^c	500	29	~ 100 ^a	12.5

^a Denotes that PPO⁻¹ resin was used.

^b Denotes that the low mol wt oligomers were extracted and that the exact SMA concentration was unknown.

^c Denotes approximation since not all of 30 phr dissolved.

RESULTS AND DISCUSSION

Series 1

PPO dissolves readily in the hot epoxy melt with time. The time needed to dissolve the PPO is less when lower mol wt PPO resin is used. The cured plaques were clear, which at first glance suggests that both PPO resins form miscible blends with epoxy. However, visual inspection of the fracture surface of broken specimens revealed that large PPO-particles were present (see Fig. 3). The clarity of the plaques must be due to the fact that the refractive indices for PPO and DGEBA-piperidine epoxy are similar.

More evidence for complete PPO phase separation was found using differential scanning calorimetry and electron microscopy. DSC results show that the glass transition temperature of the epoxy matrix was unaffected by the addition of PPO (see Table III). Inspection of the fracture surface using SEM corroborates this finding since both large and small particulate matter are observed. After soaking the fracture surface in methylene chloride and inspecting it in the SEM, we found that the particulate phase had dissolved, which indicated a lack of significant crosslinks in the PPO. Thus, it is safe to conclude that these particles consist of noncross-linked PPO. Interestingly, the large, heterogeneous

particles of PPO actually contain epoxy inclusions that are interconnected (see Fig. 4).

The existence of small homogeneous and large

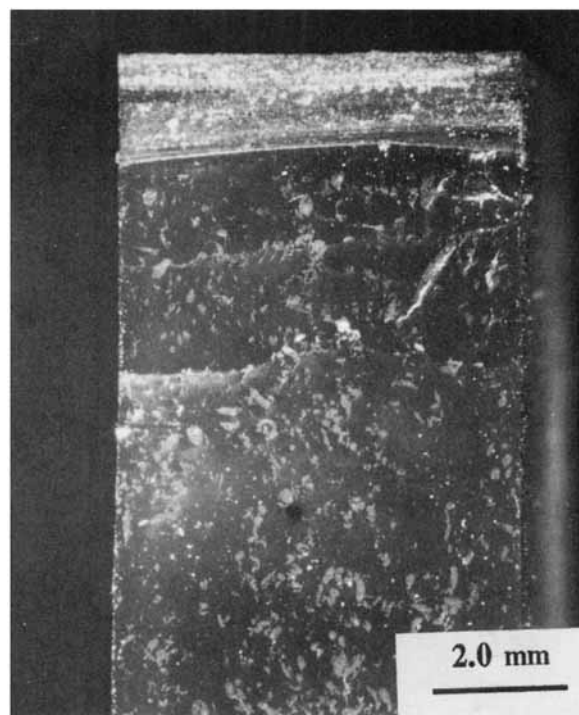


Figure 3 Optical micrograph of fracture surface of a DGEBA/Pip/PPO(10).

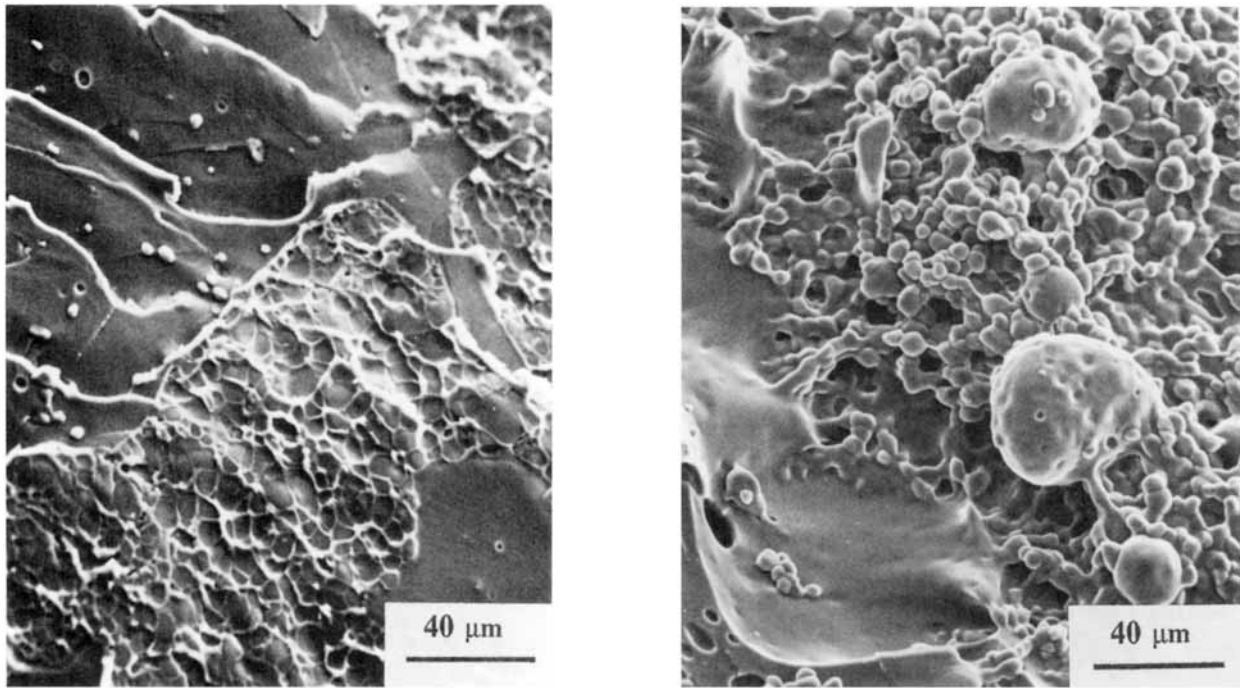


Figure 4 SEM micrograph of the fracture surface of a DGEBA/Pip/PPO(10). Note the presence of small 2-micron PPO particles as well as large PPO particles. Treating the surface with methylene chloride revealed that the PPO large particles contained epoxy domains that are interconnected.

heterogeneous PPO particles are corroborated by TEM experiments. Ultra-microtomed thin sections of this version of PPO-modified epoxy were stained with RuO_4 and were examined using TEM (see Fig.

5). These micrographs show a fine dispersed phase of PPO, together with some larger heterogeneous PPO particles containing occlusions of smaller epoxy particles. Because this system was inherently

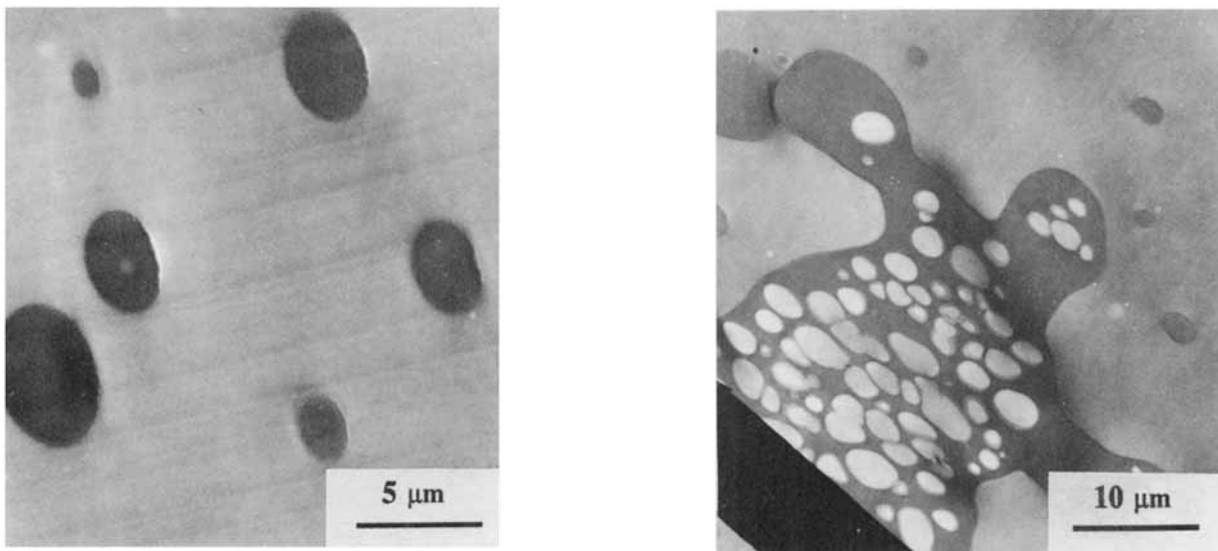


Figure 5 TEM micrographs of DGEBA/Pip/PPO(10) stained with RuO_2 . (a) shows the presence of small 2 micron PPO particles; (b) shows that the large PPO particles contain epoxy inclusions.

unstable, the specimens used for the TEM contained smaller occluded particles than those used for the SEM studies.

In order to reduce the presence of these large agglomerated particles, a lower mol wt PPO resin was tried. The lower mol wt PPO dissolved more readily into the hot, liquid epoxy. However, large heterogeneous PPO particles were still found. The elimination of these large particles required the implementation of another method. Therefore several styrene-maleic anhydride (SMA) block copolymers were tried as possible surfactants for PPO-modified epoxies in the next series. The lower mol wt PPO resin was used for the remainder of this work since it was easier to process.

Series 2

The emulsification of immiscible polymer blends by the use of block copolymers has been reported by a number of investigators.¹⁵⁻¹⁷ Sjoerdsma et al.¹⁵ have shown that the addition of a small amount of an hydrogenated PS-PB diblock copolymer in an immiscible polystyrene/polyethylene blend could improve the adhesion between the phases, resulting in enhanced mechanical properties. Welander and Rigdahl¹⁶ used an hydrogenated SBS triblock copolymer to reduce the particle size in a similar polystyrene/polyethylene blend. The emulsifying agent commonly consisted of blocks containing chemical structures identical to those which required emulsification. However, the work by Fayt et al.¹⁷ has shown that the use of diblock copolymers containing blocks that are structurally different, yet chemically similar, may be more efficient emulsifiers as long as these chemically similar blocks are soluble in the polymers to be emulsified.

To apply this same emulsification scheme to thermoplastic-modified epoxies, several styrene-maleic anhydride block copolymers were evaluated. The anhydride chemically reacted with the epoxy, thus guaranteeing miscibility.¹⁸ Polystyrene has been shown to be miscible in PPO by a number of authors.¹⁹⁻²¹ Therefore, the addition of SMA copolymers to Epoxy/PPO blends should result in a more uniform and a finer morphology.

Three different low mol wt SMA resins were evaluated as possible surfactants for PPO-modified epoxies. Unfortunately, inspection of the fracture surface revealed that the large PPO particles were still present. The number of large particles appeared to be the lowest for the formulation containing SMA-3, thus several other loadings of SMA-3 were tried. However, the sample containing the 10 phr

SMA-3 gelled prematurely. The reason for the inability of these SMA block copolymers to act as surfactants is most likely their low mol wt, which renders them unable to reduce the surface tension of the particulate phases. Therefore, this approach was abandoned and higher mol wt SMA resins were tried in the next series.

Series 3

Three higher mol wt SMA copolymers were evaluated as possible surfactants for PPO-modified epoxies. At 5 phr SMA, visual inspection suggested a greater degree of compatibilization than any of the materials in the previous series. However, the SMA pellets did not completely dissolve. We suspected that this may have been due to the presence of insoluble high mol wt fractions. Consequently, a scheme was employed to extract the soluble oligomers. SMA resin was added to a jar of epoxy and this mixture was stirred for several hours at 40-60°C. Unfortunately, the mechanical properties of the resultant blends were not consistent and the reactive nature of the two SMA copolymers caused sporadic premature gelation. Therefore, the use of SMA-4 and SMA-5 was abandoned and the remainder of this investigation focuses on SMA-6.

The SMA-6 copolymer had a higher polystyrene content than any of the previous SMA copolymers. The SMA-6 pellets completely dissolved at 5 phr loading and proved to be the best surfactant for PPO-modified epoxies. The morphology appeared much improved. SEM examination revealed that even though there were a few large particles, they were still smaller than those seen before. In an effort to completely rid this system of large PPO particles, a lower mol wt DGEBA resin was tried in Series 4.

Series 4

In retrospect, the influence of the mol wt of the epoxy resin on PPO miscibility was initially underestimated. The article by Verchere et al.²² showed that for CTBN modified epoxies, the cloud point temperature was extremely sensitive to the epoxide equivalent weight. Our crude measurements of the cloud point temperature of PPO modified epoxies revealed a 30°C increase when the epoxide mol wt was increased from 348 to 480. In fact, when the mol wt of the epoxide resin was increased to 480 g/mole, the PPO was not miscible, even at 250°C. The use of the lowest mol wt DGEBA resin, that is, Dow's DER 332 resin, in combination with the SMA-6 copolymer, resulted in a compatibilized particulate

morphology. The next series evaluates the influence of PPO content on this stabilized morphology.

Series 5

Encouraged by the elimination of large particles in the 10 phr PPO epoxy, a series of materials with varying PPO contents was made, namely 5, 10, 15, and 20 phr PPO. The morphologies of these blends are shown in Figure 6. Uniform morphologies are seen for 5, 10, and ~ 20 phr PPO, but in the 20 phr PPO blend, the PPO phase is not particulate but cocontinuous. Large PPO particles with occlusions can be seen in the material containing 15 phr PPO, but there were also regions with small 2 micron particles. Therefore, at this PPO content, there is a transition from a particulate morphology containing discrete PPO particles to a commingled morphology, consisting of cocontinuous domains of PPO and epoxy.

SUMMARY AND DISCUSSION

The series of experiments leading to the development of PPO-modified epoxies has provided the following observations (also see Table IV):

1. The addition of 10 phr high mol wt PPO to the DGEBA-piperidine epoxy system leads to a two-phase solid, consisting of small, 2 micron particles and large, 1 mm particles. Also, the large particles are heterogeneous in composition and consist of cocontinuous domains of PPO and epoxy.
2. Decreasing the mol wt of the PPO from 42,500 g/mole to 34,000 g/mole does not eliminate the occurrence of large particles.
3. The addition of low mol wt (< 4000 g/mole) SMA copolymers also does not improve the uniformity of these two-phase materials.
4. In contrast, the addition of a small amount

Table IV Summary of Observations for Series 1, 2, and 3

Designation	T_g (C)	Comments
Series 1: The Effect of PPO Mol Wt		
DGEBA/PIP	82.3	Unmodified epoxy (Control)
DGEBA/PIP/PPO-1 (10)	81.2	PPO precipitates a few large particles
DGEBA/PIP/PPO-2 (10)	82.0	PPO precipitates many large particles
Series 2: The Use of Low MW SMA Copolymers		
DGEBA/PIP	82.3	Unmodified epoxy (Control)
DGEBA/PIP/PPO (10)	82.0	PPO precipitates a few large particles
DGEBA/PIP/PPO (10)/SMA-1 (1)	85.4	Nonuniform morphology
DGEBA/PIP/PPO (10)/SMA-2 (1)	81.5	Nonuniform morphology
DGEBA/PIP/PPO (10)/SMA-3 (1)	81.4	Somewhat better uniformity
DGEBA/PIP/PPO (10)/SMA-3 (5)	84.6	Still not uniform
DGEBA/PIP/PPO (10)/SMA-3 (10)	— ^a	Gelled prematurely; approach abandoned
Series 3: The Use of High Mol Wt SMA Copolymers		
DGEBA/PIP	82.3	Unmodified epoxy (Control)
DGEBA/PIP/SMA-4 (5)	— ^a	Gelled prematurely
DGEBA/PIP/SMA-5 (5)	— ^a	Gelled prematurely
DGEBA/PIP/SMA-6 (5)	81.8	Dissolves completely
DGEBA/PIP/PPO (10)/SMA-4 (5)	— ^a	Pellets do not dissolve; smooth meniscus
DGEBA/PIP/PPO (10)/SMA-5 (5)	— ^a	Pellets do not dissolve; smooth meniscus
DGEBA/PIP/PPO (10)/SMA-6 (5)	80.9	Smooth meniscus; morphology still not uniform
DGEBA/PIP/SMA-5 (P) ^b	80.9	Epoxy with SMA oligomers (Control)
DGEBA/PIP/PPO (10)/SMA-5 (P) ^b	81.7	Smooth meniscus; morphology still not uniform
DGEBA/PIP/PPO (10)/SMA-4 (P) ^b	— ^a	Gelled prematurely; approach abandoned

^a (—) Data not available.

^b Low mol wt oligomers were extracted and the exact SMA concentration is unknown.

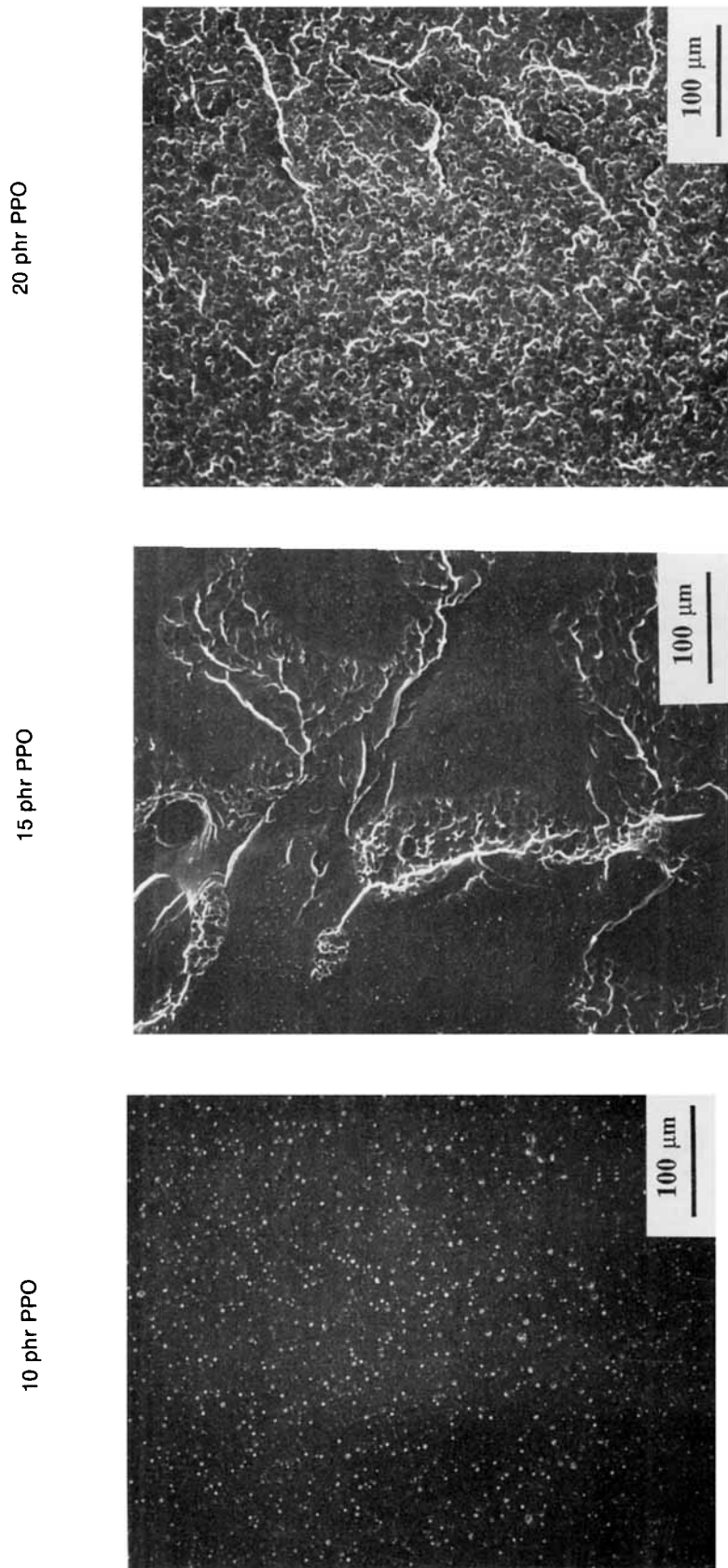


Figure 6 SEM micrographs of the fracture surfaces for a series of DGEBA/Pip/PPO(10)/SMA(1) resins. Note the uniform morphologies in the 5, 10, and 20 phr formulations. At 20 phr PPO, the morphology consisted of cocontinuous domains and the 15 phr displayed an intermediate morphology.

of a particular styrene-maleic anhydride copolymer to the DGEBA/PIP epoxy system can eliminate the occurrence of the large PPO particles in these blends. The styrene-maleic anhydride ratio of the copolymer was shown to be important.

5. The effectiveness of SMA-6 to eliminate the occurrence of large, occluded PPO particles is a function of PPO content. At 15 phr PPO there are signs of a transition in morphology and at 20 phr the morphology consists entirely of cocontinuous domains. It should be mentioned that no synthetic effort has been made on our part to tailor the structure of the SMA copolymer. Presumably, a more efficient compatibilizing agent could be made.

The use of a compatibilizing agent in thermoplastic-modified epoxies has not been previously reported in the literature. The results from this work should encourage the use of compatibilizers in similar systems and increase the number of useful blends. Compatibilized blends in thermoplastics materials are common and the transfer of this technology to thermoplastic-modified thermosets should increase the number of useful blends.

The authors wish to acknowledge the generosity of Dr. P. Yang of the Dow Chemical Co. for providing the epoxy resin, to thank Mr. Jack Huang for the TEM support, and to thank Prof. Jin of the Institute of Chemistry, Academia Sinica, Peking for the chemical analysis. This work was funded by a Materials Research Group grant from the National Science Foundation (DMR-8708405).

REFERENCES

1. C. B. Bucknall and I. K. Partridge, *Polymer*, **24**, 639 (1983).
2. R. S. Raghava, *SAMPE Symp.*, **28**, 367 (1983).
3. J. L. Hendrick, I. Yilgor, G. L. Wilkes, and J. E. McGrath, *Polym. Bull.*, **13**, 201 (1985).
4. C. B. Bucknall and I. K. Partridge, *Polym. Eng. Sci.*, **26**, 54 (1986).
5. J. A. Cecere and J. E. McGrath, *Polym. Preprts.*, **27**, 299 (1986).
6. H. Jabloner, B. J. Swetlin, and S. G. Chu, U.S. Patent No. 4,656,207 (1987).
7. S. G. Chu, B. J. Swetlin, and H. Jabloner, U.S. Patent No. 4,656,208 (1987).
8. R. S. Raghava, *J. Polym. Sci. B Polym. Phys.*, **25**, 1017 (1987).
9. R. S. Raghava, *J. Polym. Sci. B Polym. Phys.*, **26**, 65 (1988).
10. S. C. Kim and H. R. Brown, *J. Mater. Sci.*, **22**, 2589 (1987).
11. H. J. Sue, Ph.D. thesis, University of Michigan, 1988.
12. Z. Fu and Y. Sun, *Polym. Preprts.*, **29**, 177 (1988).
13. C. B. Bucknall and A. H. Gilbert, *Polymer*, **30**, 213 (1989).
14. R. A. Pearson and A. F. Yee, *Polymer*, to appear.
15. S. D. Sjoerdsma, C. A. M. Bleijenberg, and D. Heikens, *Polymer*, **22**, 619 (1981).
16. M. Welander and M. Rigdahl, *Polymer*, **30**, 207 (1989).
17. R. Fayt, R. Jerome, and Ph. Tessie, *Polym. Eng. Sci.*, **27**, 328 (1987).
18. H. Lee and K. Neville, *Handbook of Epoxy Resins*, McGraw-Hill, New York, 1967.
19. H. E. Bair, *Polym. Eng. Sci.*, **10**, 247 (1970).
20. A. R. Schultz and B. M. Gendron, *J. Appl. Polym. Sci.*, **16**, 461 (1972).
21. W. J. MacKnight, F. E. Karasz, and J. R. Fried, in *Polymer Blends*, Paul and Newman, Eds., Academic, New York, 1978, Vol. 1, Chap. 5.
22. D. Verchere, H. Sautereau, J. P. Pascault, S. M. Moschiar, C. C. Riccardi, and R. J. J. Williams, *Polymer*, **30**, 107 (1989).

Received June 24, 1992

Accepted July 27, 1992