

Toughening of Epoxy Resins by *N*-Phenylmaleimide–*N*-Cyclohexylmaleimide–Styrene Terpolymers

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SYNOPSIS

N-Phenylmaleimide (PMI)–*N*-cyclohexylmaleimide (CMI)–styrene (St) terpolymers were used to improve the toughness of bisphenol-A diglycidyl ether epoxy resin cured with *p,p'*-diaminodiphenyl sulfone (DDS). The terpolymers were effective as modifiers for toughening of the epoxy resin system. The most suitable compositions for the modification of the epoxy resin were inclusion of 10 wt % of the terpolymer (45 mol % PMI, 5 mol % CMI, and 50 mol % St units) with M_w higher than 247,000, which led to ca. a 140% increase in the fracture toughness (K_{IC}) of the cured resin with a medium expense of mechanical properties. The glass transition temperatures of the modified resins were equal to or higher than that of the parent epoxy resin. The morphologies of the modified resins were dependent on the terpolymer molecular weight and concentration. On addition of 5 wt % of the terpolymer (M_w 400,000), the modified resins had two-phase morphologies and the terpolymer-rich dispersed particles in the epoxy matrix. On addition of more than 8 wt % of the terpolymer, the morphologies of the cured resins changed drastically and showed a tendency to form cocontinuous phases. The toughening mechanism was discussed in terms of the morphological behaviors of the modified epoxy resin systems. © 1993 John Wiley & Sons, Inc.

INTRODUCTION

Epoxy resins are one of the most important thermosetting polymers and have wide use as structural adhesives and matrix resins for fiber composites, but their cured resins have one drawback: They are brittle and have poor resistance to crack propagation. The toughness of bifunctional epoxy resins such as diglycidyl ether of bisphenol A (DGEBA) has been increased by blending with reactive liquid rubbers such as carboxyl-terminated butadiene acrylonitrile rubbers (CTBN)^{1,2} or epoxide-containing acrylic rubbers.^{3–7} These elastomers, however, are not always effective modifiers for highly cross-linked epoxy matrix.

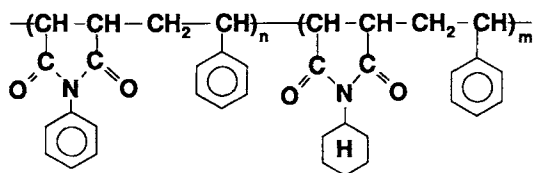
Engineering thermoplastics are interesting materials as modifiers for epoxy resins from the viewpoint of the maintenance of mechanical and thermal properties for the matrix resins. Modifications of

epoxy resins with various types of ductile thermoplastics have been studied as an alternative to reactive rubber toughening for improving the toughness of epoxy resins. At first, commercial poly(ether sulfone)s (PESs) such as Vitrex were used as modifiers in modification of polyfunctional epoxies such as tetraglycidyl diaminodiphenylmethane (TGDDM), but less effective owing both to their high cross-linking structure and to poor interfacial bonding between two uncompatibilized phase in the cured resin.^{8–10} Terminally functionalized polysulfones are more effective modifiers than is commercial PES.^{11,12} Recently, other engineering plastics have also been examined as modifiers, where polyetherimides,^{13–16} poly(aryl ether ketone)s,^{17,18} and poly(phenylene oxide)¹⁹ have been reported as effective modifiers. Aromatic polyesters, prepared by the reaction of (iso)phalic acids and α,ω -alkanediols, were also effective modifiers.²⁰ Most recently, we found that poly(*N*-phenylmaleimide-*alt*-styrene) (PMS) was an effective modifier for the DGEBA epoxy resins.²¹ In spite of the carbon–carbon single-bond repeating units, PMS has a high T_g of over

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200°C. It is thought that the bulkiness of two side groups in PMS prevents the molecular motion of the main chain, which raises the T_g of PMS. Furthermore, the high molecular weight polymer could be easily prepared, whereas it is rather difficult to obtain engineering thermoplastics with high molecular weights, as the thermoplastics are generally prepared by polycondensation.

In this study, poly(*N*-cyclohexylmaleimide-*alt*-styrene) was at first prepared, but the copolymer could not be used as a modifier because of its poor compatibility with the DGEBA/*p,p'*-diaminodiphenyl sulfone (DDS) matrix. Then, *N*-phenylmaleimide (PMI)-*N*-cyclohexylmaleimide (CMI)-styrene (St) terpolymer (PCMS) was used, because it was expected that the morphologies of the cured resins could be controlled by PCMS composition and concentration and that such an approach would give some information on the relationship between the effectiveness as modifiers for toughening of the epoxy resin and the molecular structure of PMS-like thermoplastics:



PCMS

Scheme 1

EXPERIMENTAL

Materials

The epoxy resin used was the liquid bisphenol A diglycidyl ether (DGEBA) (Epikote 828, Shell Chemical Industrial Co., epoxy equivalent weight (EEW) 190). DDS was used as the curing agent. PMI and CMI were provided by Nippon Shokubai Corporation. St was purified in the usual way. Other reagents were used as received.

Measurements

Molecular weights of terpolymers were determined by gel permeation chromatography (Shimadzu LC-5A instrument) using polystyrene standards. Mechanical properties of cured resins were measured with a Shimadzu autograph AGS-500B universal testing machine. Flexural tests were carried out at a crosshead speed of 2 mm/min (JIS K7203). Fracture toughness, K_{IC} , was measured in a three-point

bent geometry at a crosshead speed of 1 mm/min (ASTM E-399). Glass transition temperatures of both terpolymers and cured resins were measured by differential scanning calorimetry (Shimadzu DSC 41 M type) at a heating speed of 10°C/min. Scanning electron micrographs were taken with a Hitachi SEM S-2100A instrument using failed specimens in the K_{IC} tests. Dynamic viscoelastic analysis was performed with a Rheometrics RDS-II type (Rheometrics Co.) between -150 and 250°C at a heating speed of 5°C/min at frequency of 1 Hz.

Preparation of *N*-Phenylmaleimide-*N*-Cyclohexylmaleimide-Styrene Terpolymers

The terpolymers were prepared in the 1 : 1 mol ratio of *N*-substituted maleimides and St. The molecular weight of PCMS was controlled using dodecanethiol as a chain transfer agent. A typical procedure is as follows:

A flask was charged with PMI (39.0 g, 0.225 mol), CMI (4.48 g, 0.025 mol), St (26.0 g, 0.25 mol), acetone (500 mL), azobisisobutyronitrile (AIBN, 0.82 g, 1 mol %), and dodecanethiol (0.05 g, 0.05 mol %). The flask was purged with N_2 for 0.5 h and a N_2 atmosphere was maintained throughout the polymerization. The polymerization mixture was stirred at 60°C for 3.5 h. After the polymerization was over, the polymerization mixtures were diluted with tetrahydrofuran (THF). Then, the terpolymer was isolated using methanol as precipitant and purified twice by reprecipitation with THF/methanol. The white solid polymer was dried *in vacuo* at 60°C and obtained in a quantitative yield.

Curing Procedure

The terpolymer was dissolved into the epoxy resin without solvents by heating at 140 or 160°C, dependent on the molecular weight of PCMS. Then, the curing agent, DDS, was added into the mixture, which was kept at 120°C for about 1 h to dissolve the DDS. The resulting clean mixture was poured into a silicone mold preheated at 100°C. The curing cycle was 120°C/1 h + 180°C/5 h. The curing agent was used stoichiometrically to the epoxy resin. The amount (wt %) of PCMS used was calculated based on the epoxy resin.

RESULTS

Characterization of Terpolymers

It has been reported that the copolymerization of PMI and St gives alternating copolymers, indepen-

Table I Characterization of *N*-substituted Maleimide–Styrene Terpolymers^a

Entry No.	Feed Composition			C ₁₂ H ₂₅ SH ^b (mol %)	Yield (%)	M _n ^c (10 ⁴)	M _w ^d (10 ⁴)	M _w /M _n	T _g ^e (°C)
	PMI	CMI	St						
PCMS I series									
1 ^e	45	5	50	0.00	98.0	2.1	12.2	5.91	221
4				0.33	92.0	4.0	10.1	2.53	217
5				0.10	94.0	7.4	18.7	2.54	216
6				0.05	93.0	9.9	24.7	2.48	222
7				0.02	92.0	15.8	35.8	2.26	223
3				0.00	92.0	19.4	40.2	2.07	223
PCMS II series									
2 ^e	40	10	50	0.00	99.5	2.7	14.1	5.30	213
8				0.05	91.0	8.2	19.7	2.40	220

^a AIBN 1 mol %; in acetone at 60°C.

^b Chain transfer agent.

^c By GPC.

^d By DSC.

^e In toluene at 70°C.

dent of the feed monomer composition.²² The copolymerization of CMI and St gave also an alternating copolymer.²³ The terpolymerization of PMI, CMI, and St must also afford alternating copolymers composed of PMI/St and CMI/St units. Table I reports some characteristic properties of the terpolymers. In this study, the terpolymer composition is expressed conveniently in terms of the feed monomer composition. The glass transition tem-

perature of terpolymer PCMS was over the range of 216–223°C.

Mechanical and Thermal Properties of Modified Epoxy Resins

Table II shows representative results for the modification of the epoxy resin with PCMS I (45 mol % PMI, 5 mol % CMI, and St 50 mol % units) or

Table II Physical Properties of Cured Epoxy Resins

PCMS ^a Entry No.	Content Wt %	K _{IC}		Flexural Properties (kgf/mm ²)			T _g ^c (°C)
		(MN m ^{2/3})	n ^b	Strength	Modulus	n ^b	
	0	0.71 ± 0.02	6	16.6 ± 0.4	337 ± 8	8	180
PCMS I (PMI : CMI : St, 45 : 5 : 50) series							
4	5	0.96 ± 0.04	6	17.1 ± 0.5	383 ± 6	7	182
4	10	0.99 ± 0.01	6	15.0 ± 0.9	384 ± 6	6	186
7	5	0.95 ± 0.04	7	15.4 ± 1.6	377 ± 12	7	186
7	10	1.69 ± 0.05	6	9.8 ± 0.4	377 ± 6	6	186
PCMS II (PMI : CMI : St, 40 : 10 : 50) series							
2	5	0.93 ± 0.04	7	16.0 ± 0.4	310 ± 6	6	176
2	10	1.00 ± 0.04	6	15.9 ± 0.6	334 ± 8	7	176
8	5	0.95 ± 0.03	7	17.3 ± 0.7	385 ± 5	6	184
8	10	1.53 ± 0.04	7	8.7 ± 0.5	395 ± 4	7	186

^a Entry number in Table I.

^b Specimen number.

^c By DSC.

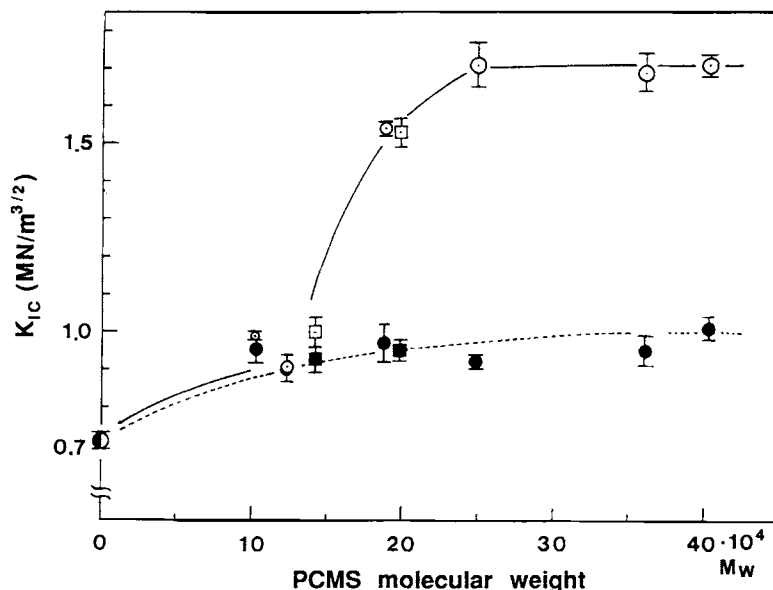


Figure 1 Dependence of fracture toughness for modified resins on PCMS molecular weight. PCMS I: (●) 5 wt % addition; (○) 10 wt % addition. PCMS II: (■) 5 wt % addition; (□) 10 wt % addition.

II (40 mol % PMI, 10 mol % CMI, and St 50 mol % units). The cured parent epoxy resin was transparent, but the modified resins became translucent. In the modification with PCMS I of lower molecular weight (M_w) than M_w 150,000, the fracture toughness, K_{IC} , increased 30–40% at no expense of mechanical and thermal properties of the modified resin on addition of 5–10 wt % of PMS. On addition of 15 wt % of PCMS (M_w 122,000), the viscosity of the uncured epoxy mixture at 120°C increased greatly and the cured resins could not be obtained. The use of 10 wt % of PCMS I with M_w 187,000 led to a significant increase (120%) in K_{IC} . Figure 1 shows the PCMS MW dependence of the fracture toughness, K_{IC} , for the modified resins. On addition of 5 wt % of PCMS I, K_{IC} increased 30–40% independent of PCMS MW. On addition of 10 wt % of PCMS I, K_{IC} for the modified resins increased with increasing PCMS MW and leveled off, together with decreasing flexural strength. Inclusion of 10 wt % of PCMS I (M_w 247,000) brought about a 140% increase in K_{IC} . Flexural moduli for the PCMS I-modified resins increased, compared to that for the parent epoxy resin. The T_g values for the modified resins were equal to or higher than that for the parent epoxy resin (Table II).

Figures 2 and 3 show the mechanical and thermal properties of the PCMS I-modified resins as a function of concentration of PCMS. The K_{IC} values for the modified resins increased gradually on addition of lower MW PCMS I. In the modification with higher MW PCMS I, the increased PCMS concen-

tration led to a drastic increase in K_{IC} . The use of PCMS I with lower M_w (122,000) brought about no decrease in flexural strength for the cured resins. In the use of PCMS I with higher M_w (402,000), the flexural strength for the modified resins hardly changed on addition of 5 wt %, but decreased in the modification with more than 8 wt % of PCMS. Flexural moduli for the modified resins did not change,

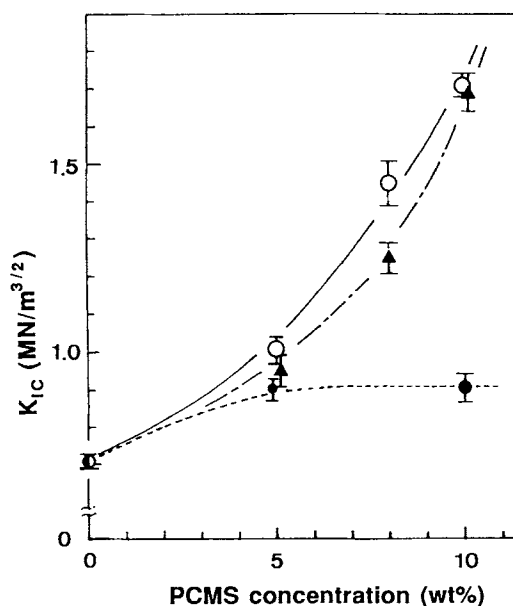


Figure 2 Fracture toughness for modified resins as function of PCMS I concentration: (○) M_w 402,000; (▲) M_w 358,000; (●) M_w 122,000.

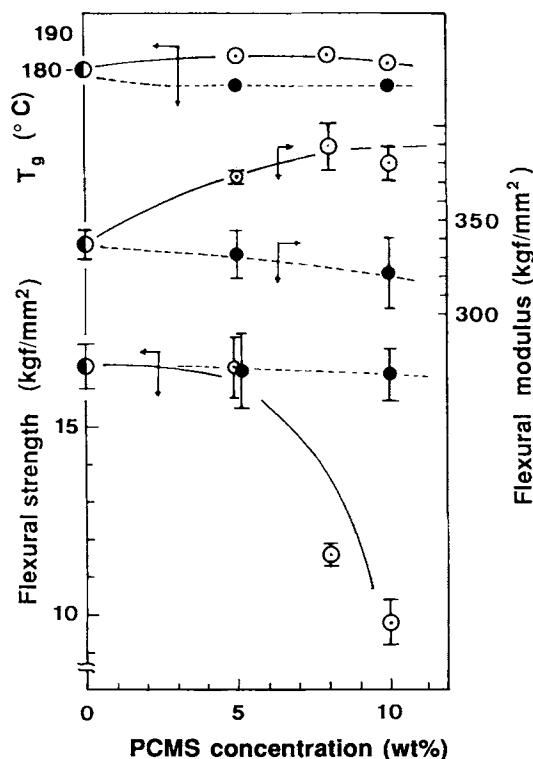


Figure 3 Physical properties of PCMS-modified resins as function of PCMS I concentration: (○) M_w 402,000; (●) M_w 122,000.

compared to that for the parent resin. T_g 's for the modified resins were almost equal to or higher than that for the unmodified resin.

In the modification with PCMS II, the modifier MW dependence of K_{IC} for the modified resins was similar to that for the PCMS I ones (Fig. 1). K_{IC} for the PCMS II (M_w 197,000)-modified resin increased 115% with a medium expense of flexural strength on addition of 10 wt %, compared to that for the parent epoxy resin. Flexural behavior for the PCMS II-modified resins was also similar to that for the PCMS I ones (Table II). T_g 's for the modified resin were almost equal to or higher than that for the parent epoxy resin. It is noteworthy that PCMS I with the highest M_w (402,000) is slightly difficult to handle, because of an increase in the viscosity of the uncured epoxy mixture.

Morphologies of Modified Epoxy Resins

The morphologies of the cured resins were investigated by scanning electron micrographs (SEMs). The parent epoxy resins had only one phase as reported previously.³ Inclusion of PCMS led to phase separation of the cured resins. The morphologies of the cured resins changed drastically, dependent on

the PCMS MW and concentration. Figure 4 shows SEMs of the cured resins modified with 10 wt % of various MW PCMS I. The use of PCMS with low M_w (122,000) led to two-phase morphology with PCMS-rich spherical particles dispersed in the epoxy-rich matrix; the volume fraction (V_f) and the average diameter (D) of the particles were, respectively, 0.06 and 0.72 μm on 5 wt % addition of PCMS I and 0.11 and 0.83 μm on 10 wt % addition. The use of 10 wt % of PCMS I with M_w 187,000 led to a drastic change in the morphology for the modified resin, which had a cocontinuous phase morphology. Figure 5 shows a change in morphological behavior with increasing PCMS I concentration in the modification with high MW PCMS. On addition of 5 wt % of PCMS I (M_w 402,000), the modified resin had a two-phase morphology with spherical particles dispersed in the matrix, where V_f and D are 0.14 and 1.64 μm , respectively. In the use of 8 wt % of the same modifier, the particles coagulated and formed the continuous phase, but a small amount of particles remained in the matrix. The morphology of the cured resin modified with 10 wt % of PCMS I (M_w 402,000) was similar to that with 8 wt % addition of the same modifier.

The morphological behavior in the modification with PCMS II was similar to that in the PCMS I modification system. In the use of lower M_w PCMS II (141,000), the modified resins had two-phase morphologies; V_f and D were, respectively, 0.12 and 0.80 μm on 5 wt % addition of PCMS II and 0.22 and 1.20 μm on 10 wt % addition. When using 10 wt % of higher M_w PCMS II (197,000), the modified resin had the cocontinuous phase morphology.

Dynamic Viscoelastic Analysis of Modified Epoxy Resins

Dynamic viscoelastic analysis can give information on the microstructure of cured resins. Figure 6 shows the storage moduli, G' , and $\tan \delta$ for the unmodified and PCMS I (M_w 247,000)-modified resins. The peak position of the α -relaxation in the $\tan \delta$ curves hardly changed in the modification with PCMS, but the peak became broader on addition of 10 wt % of PCMS, compared with dynamic viscoelastic behavior for the parent epoxy resin. The storage moduli in the glass state were comparable to the unmodified resin.

DISCUSSION

PCMS was an effective modifier for improving the toughness of the epoxy resin systems, but fractural

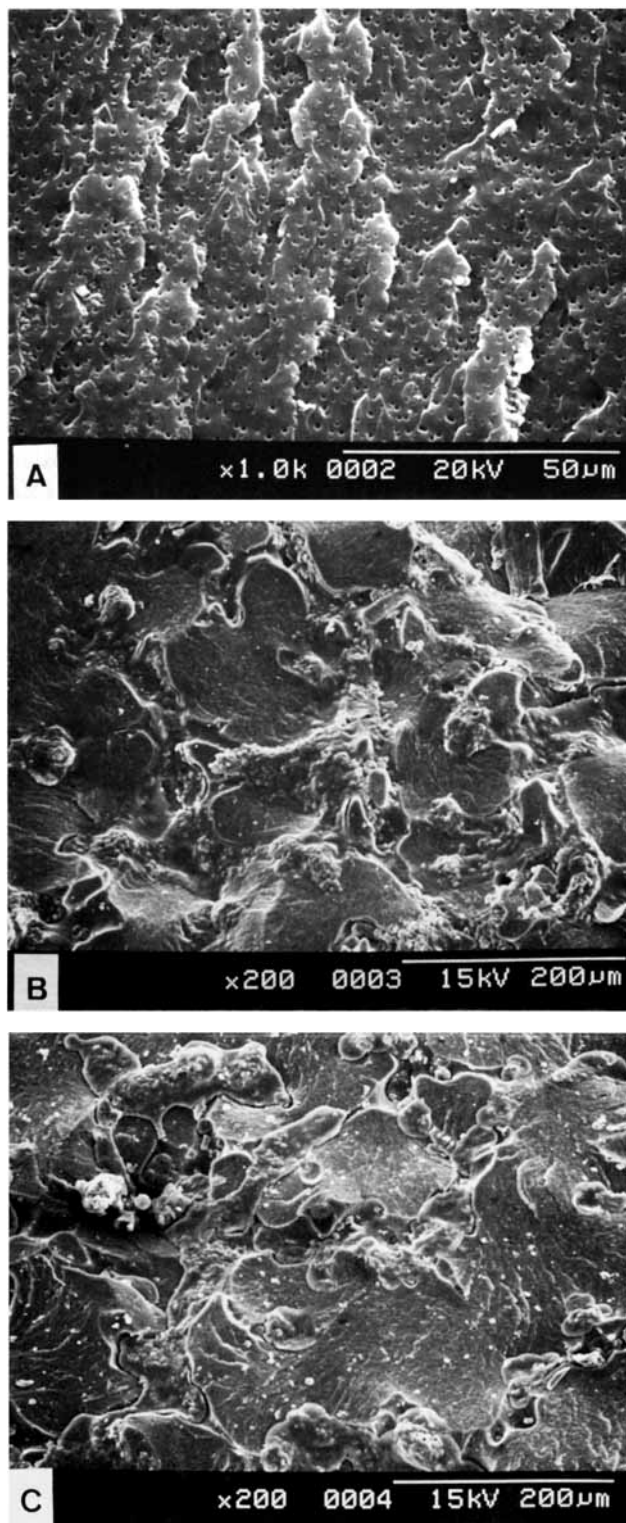


Figure 4 SEMs of fracture surfaces for the cured resins modified with 10 wt % of PCMS I. M_w of PCMS and the scale bar, in μm , respectively: (A) 122,000 and 50; (B) 187,000 and 200; (C) 247,000 and 200.

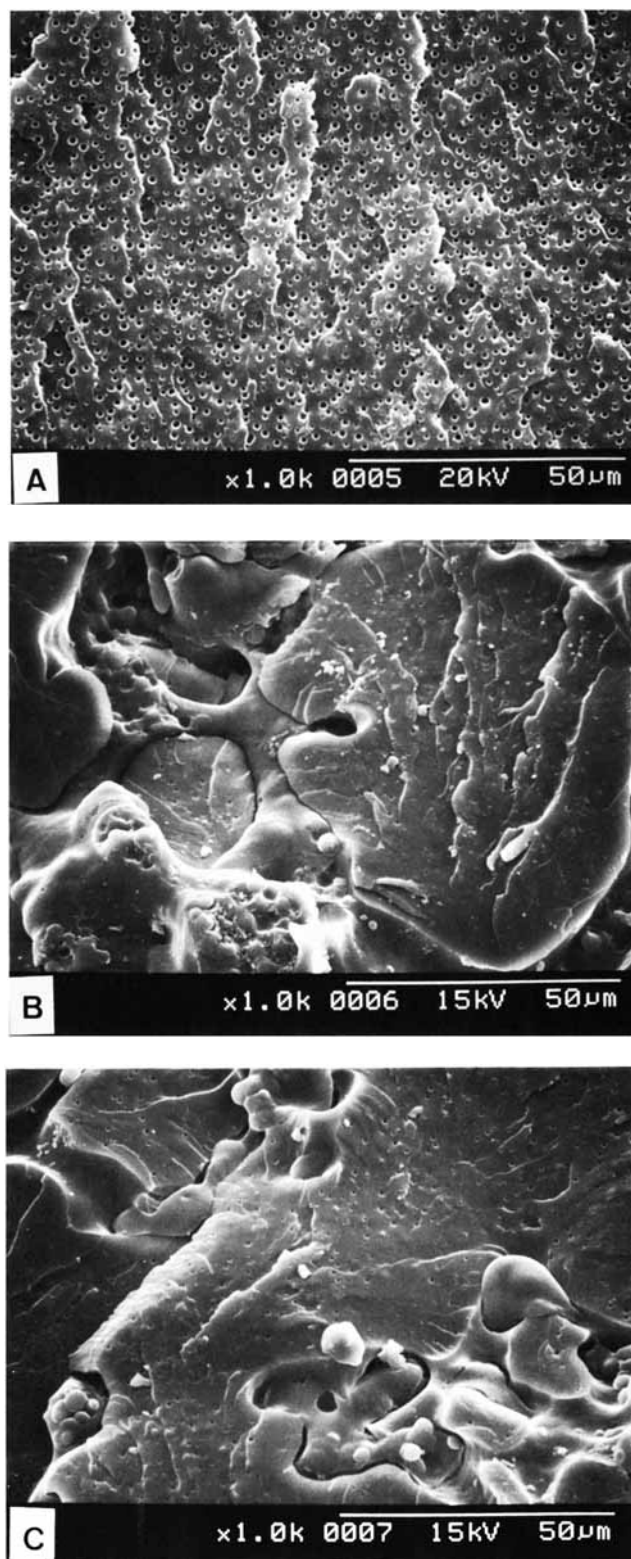


Figure 5 SEMs of fracture surfaces for the cured resins modified with PCMS I (M_w 402,000). Amount of PCMS I: (A) 5 wt %; (B) 8 wt %; C, 10 wt %. The scale bars are 50 μm .

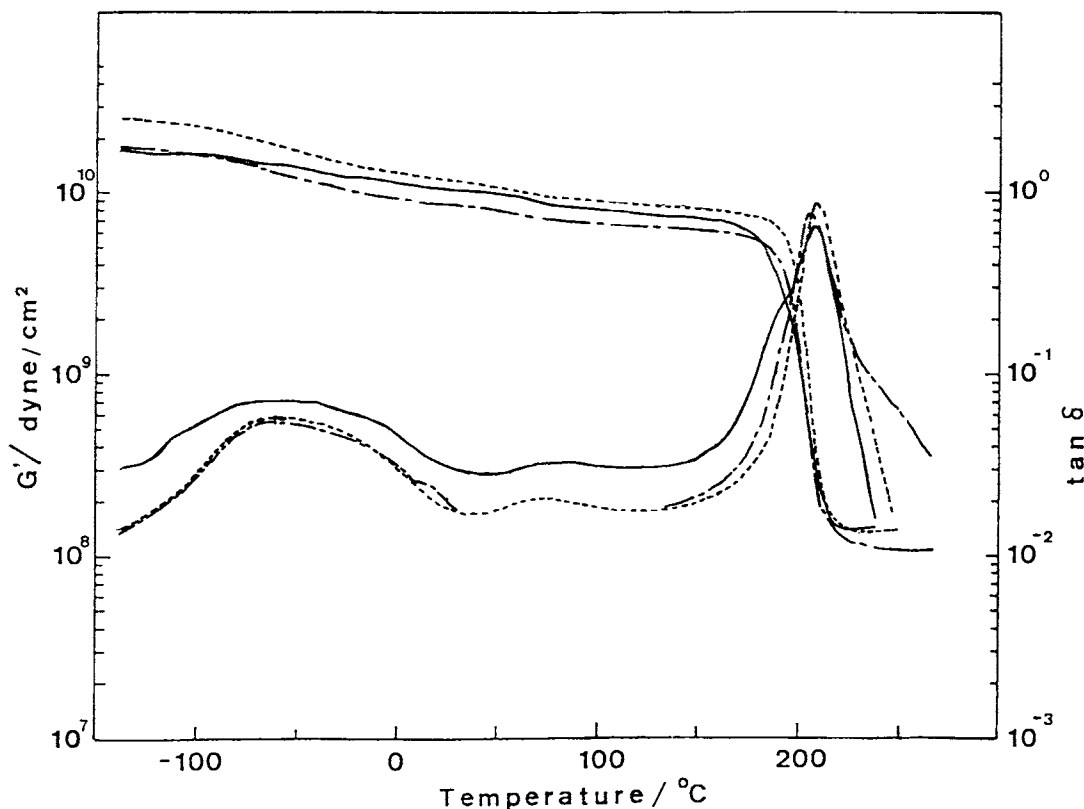


Figure 6 Dynamic viscoelastic analysis for unmodified and PCMS-modified resins. PCMS I, M_w 247,000: (—) control; (---) 5 wt % addition; (- - -) 10 wt % addition.

behavior was changed to a considerable extent, dependent on both PCMS MW and concentration. The fractural behavior was investigated in detail in the PCMS I modification system. The use of 10 wt % of PCMS I with M_w lower than 122,000 led to a medium increase in K_{IC} at no sacrifice of mechanical and thermal properties of the modified resins, and the modified resins had two-phase morphologies. In the modification with low MW PCMS I, the extent of the improvement in the toughness is comparable to that in the modification with the reactive acrylic elastomers, butyl acrylate-glycidyl methacrylate-acrylonitrile terpolymers, which were reported as effective modifiers previously.^{6,7}

In the modification with high MW PCMS I, the fractural behavior changed drastically dependent on PCMS I concentration (Figs. 1 and 2). On addition of 5 wt % of PCMS I with M_w higher than 187,000, the extent of toughening is comparable to or slightly larger than that in the use of lower MW PCMS I. K_{IC} increased significantly when using 10 wt % of higher MW PCMS I. All PCMS I with M_w higher than 247,000 used in this study have a similar efficiency as a modifier for toughening of the epoxy resin. PCMS I of M_w 247,000 is one of the most

effective modifiers and its 10 wt % inclusion leads to 140% increase in K_{IC} , whereas K_{IC} increases 30% on 5 wt % addition.

The dependence of K_{IC} on PCMS I concentration in the higher MW PCMS modification systems is quite different from that in the modification of epoxies with reactive acrylic elastomers, where K_{IC} increased gradually with increasing the elastomer concentration.³⁻⁶ In the modification with PCMS I of M_w higher than 187,000, K_{IC} increased rather discontinuously with the increase in PCMS I concentration (Fig. 2). Figure 2 also indicates that the effectiveness of PCMS II as a modifier is similar to that of PCMS I. When using the most suitable modifier, the extent of toughening in the PCMS modification system is comparable to that in the PMS one.²¹ For example, inclusion of 10 wt % modifier leads to ca. 140% increase in K_{IC} for the higher MW PCMS I-modified resins and 130% increase for the PMS (M_w 345,000)-modified resin.²¹

The fractural behavior in the modification of the epoxy resin with PCMS is slightly different from that in the PMS modification system. The main difference between both modifiers is that the most suitable MW range of PCMS is far wider than that

of PMS in the modification of the epoxy resin; the effectiveness as modifier of PCMS having M_w between ca. 250,000 and 400,000 is similar in the former modification system, but the $M_w - K_{IC}$ curve has a maximum at PMS M_w of ca. 350,000 in the latter one. This result could be explained by the difference in the compatibility of the modifiers with the epoxy matrix as shown in the morphological behavior of the modified resins.

The morphological behavior of the modified resins is interesting and instructive when considering the toughening mechanism in the present modification system. The morphologies of the modified resins changed drastically, dependent on both PCMS MW and concentration. On addition of 5 wt % of PCMS, the modified resins had two-phase morphologies with PCMS-rich spherical particles dispersed in the epoxy matrix, independent of PCMS composition and MW. Inclusion of 10 wt % of PCMS led to a drastic change in the morphology of the modified resin, dependent on PCMS MW; in the use of PCMS I with M_w less than 122,000, two-phase morphologies were observed, but PCMS particles coagulated and formed the cocontinuous phase in the modification with PCMS I of M_w higher than 187,000 (Fig. 4). The PCMS concentration dependence of the morphological behavior was investigated in the modification with high M_w (402,000) PCMS I (Fig. 5); two-phase morphologies were observed on addition of 5 wt % of the modifier, but on addition of more than 8 wt % of the same modifier, the PCMS-rich particles coagulated and formed the cocontinuous phase, together with the significant increase in K_{IC} (Fig. 3). When using PCMS II with M_w 141,000, the modified resins had the two-phase morphology, and their V_f and D were larger than those for the PCMS I (M_w 122,000) on both 5 and 10 wt % addition. The modified resin for 10 wt % of the high M_w PCMS II (197,000) had the cocontinuous morphology. These morphological behaviors correspond to the dependence of K_{IC} on PCMS M_w and concentration. The significant increase in K_{IC} for the modified resin can be attained in the modification system having the cocontinuous phase. The MW dependence of the change in the morphologies for the PCMS-modified resins is similar to that for the PMS-modification system.²¹

In the previous paper, solvent-etched fracture surfaces were taken by SEMs to examine the interfacial adhesion between the PMS-rich phase and the epoxy matrix; the PMS-rich phase was clearly removed by etching with THF in both two-phase morphologies and cocontinuous morphologies.²¹ This result indicates that there are no chemical in-

terfacial adhesion between the epoxy matrix and PMS. It is thought that the interfacial adhesion between the matrix and the PCMS-rich phases would be also based on the physical interactions, considering similar molecular structure of both modifiers. In the PCMS-modification system, flexural modulus and T_g did not decrease by modification, but the use of effective PCMS with higher MW led to a medium decrease in flexural strength, which could be explained by insufficient interfacial adhesion between the matrix and the modifier, based on physical interactions such as the van der Waals forces. Such deficiency would be overcome by using the modifier containing functional groups that are able to react with the epoxy matrix.

Dynamic viscoelastic analysis showed no evidence of phase separation in the modification of the epoxy resin with PCMS I (M_w 247,000). There was no shoulder peak in the α -relaxation of the $\tan \delta$ curve. This behavior is different from that in the PMS (M_w 344,000)-modification system;²¹ the shoulder peaks at ca. 175°–185° in the $\tan \delta$ curve were observed based on PMS-rich phases in the latter modification and their transition temperatures were lower than T_g (219°C) (by DSC) of PMS. The dynamic viscoelastic behavior in the PCMS-modification system would be explained by the incorporation of the epoxy matrix into the PCMS-rich phase, considering the morphological results for the modified resins with two-phase structure.

The fractural behavior corresponds to the morphology of the modified resin. In every case of this study, the toughness of the epoxy resin could be improved significantly because of the cocontinuous structure of the modified resins. Similar behavior was observed in the PMS-modification system. These results suggest that two toughening mechanisms could operate in the present modification system as well as in the modification of epoxies with PMS; one mechanism is due to the two-phase separation structure and similar to that in the rubber-modification systems,¹ and the other mechanism is based on the cocontinuous phase morphology. The latter mechanism would be similar to that in the modification of epoxies with engineering thermoplastics such as polyetherimides^{13,15} and poly(ether ketone)s.¹⁸

In the modification of tetraglycigyl-4,4'-diaminodiphenyl methane with polyetherimides (Utem 1000, GE) (PEI), K_{IC} for the cured resin modified with 25 phr of PEI increased 180%, whereas the phase inversion of morphologies was observed and the toughening of epoxies could be attained by the absorption of fractural energy due to ductile drawing

and tearing of the thermoplastic continuous phase. We have also found that poly(phthaloyl diphenyl ether)s (PPDE) were effective modifiers for DGEBA resins cured with methyl hexahydrophthalic anhydride and that the cocontinuous structure due to the spinodal decomposition was formed in the modified resin with PPDE of higher MW (M_{GPC} 56,300), together with a significant increase in K_{IC} .¹⁸

Engineering thermoplastics have high T_g 's based on their rigid structures. PCMS also has high T_g as well as does PMS, because of the retardation of molecular motion of the main chains based on the bulky side chains. One of the drawbacks in the use of the conventional engineering thermoplastics as modifiers is poor processability, which is based mainly on poor compatibility with the uncured epoxy resin; in general, the use of a small amount of solvent is essential to dissolve the thermoplastics into epoxies in the modification with the thermoplastics. All PCMS used in this study are soluble in the epoxy resin without using solvent, and PCMS I of M_w higher than 247,000 are comparably effective modifiers to the engineering thermoplastics such as polyetherimides. The effectiveness of PCMS as a modifier could be explained by its similar structure to PEI, where the polar imide units in PEI and PCMS would play an important role in the physical interfacial adhesion between the epoxy matrix and the thermoplastics. The T_g values for the modified resins with PCMS are almost equal to or higher than that for the unmodified epoxy resin.

In conclusion, PCMS is an interesting and effective modifier for improving the toughness of epoxy resins. Two toughening mechanisms operate in the present modification system dependent on PCMS MW and concentration: One mechanism is due to the two-phase separation structure, and the other, to the cocontinuous phase morphology. The most effective modification for the cured DGEBA resin can be attained by the latter phase structure. The suitable compositions for modification of the epoxy resin were inclusion of 10 wt % of PCMS I with M_w higher than 247,000. The drawback in the present modification system is the medium decrease in the flexural strength, perhaps owing to rather poor interfacial adhesion between the epoxy matrix and PCMS. It is thought that the efficiency of the modifier would increase by enhancing interfacial adhesion due to chemical bonding.

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