

Photosynthesis and Application of Polyfunctional Poly(*n*-Butyl Acrylate) Elastomers for Use in Epoxy Resin Toughening

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SYNOPSIS

Medium molecular weight, novel polyfunctional elastomers, namely epoxy groups on poly(*n*-butylacrylate) (ETPnBA) and carboxyl groups on poly(*n*-butylacrylate) (CTPnBA) were photosynthesized for evaluation as the toughening agents in epoxy resins. The effect of the functionality and kind of functional group of the elastomers upon the toughening of epoxy resins modified with these rubbery copolymers as a second phase was investigated by tensile tests, impact test, and electron microscopy. It was found that there exists an optimum functionality of elastomers for maximum impact resistance in epoxy groups (ETPnBA) and carboxyl groups (CTPnBA) copolymer-modified systems. Studies on morphology of the modified epoxy resin system indicated that the better toughening effects of epoxy groups ETPnBA in an epoxy resin matrix can be attributable to the existence of a multiple distribution of particle sizes. The aggregation of rubber particles occurring in carboxyl group CTPnBA modified epoxy resin caused a loss of toughness.

INTRODUCTION

The incorporation of elastomers into epoxy resins has been an active area of research over the past two decades; the primary reason is the improvement of toughness of the modified materials. One approach is based on the incorporation of a small proportion of a reactive liquid polymer (RLP) into the epoxy matrix, typically between 10 and 20%.¹

It is well known that, for effective toughening of epoxy resin, the degree of rubber-epoxy interfacial bonding is an important factor. It has been suggested that various reactive groups, such as carboxyl, hydroxyl, epoxy, mercaptan, and amine, may be used for chemical bonding. The reactive groups are present randomly on the rubber molecule backbone or located at the ends of the polymeric chain. McGarry and Willner showed that the carboxyl-terminated copolymer of butadiene and acrylonitrile (CTBN)

is useful for the improvement of the fracture toughness of epoxy resin.² Cazit and Bell^{3,4} reported that in both carboxyl-terminated poly(butylacrylate) liquid rubber (CTPnBA) and *n*-butyl acrylate/acrylic acid (nBA/AA) copolymer, the effective toughening depends on effective bonding between the reactive rubber and epoxy matrix. A family of nBA/AA copolymers with functionality of 1.62–9.93 was introduced by Lee et al.,⁵ and the results indicated that the improvement of adhesion strength could be achieved by incorporating the nBA/AA copolymer in DGEBA epoxy matrix and that an optimum functionality for matrix adhesion strength exists.

We attempt to investigate the effect of the kind of functional groups, their site on the polymeric chain, and the functionality of the rubbery modifiers in the toughening of epoxy resin. The polyfunctional poly(*n*-butylacrylates) reported in this study fall into two categories: epoxy functional poly(*n*-butylacrylate) (ETPnBA) and carboxyl functional poly(*n*-butylacrylate) (CTPnBA). These novel rubbery polymers for modified epoxy resin have been specifically investigated for mechanical properties and morphologies here.

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EXPERIMENTAL

Materials

n-Butylacrylate (nBA), acrylate acid (AA) (Shanghai No. 1 Chemical Reagent Factory), and glycidylmethacrylate (GMA) (Aldrich Chemical Co.) were distilled twice under reduced pressure (about 20 mm Hg) immediately before use. Benzophenone (BP) (Shanghai No. 1 Chemical Reagent Factory) was recrystallized twice from ethanol-water (1 : 1 by volume) and dried for 1 day under 0.01 mm Hg at 60°C. Dithiodipropionic acid (DTPA) (Aldrich Chemical Co.) was used as received. Solvents such as benzene, acetone, chloroform, methanol, and dioxane were used after dried and distillation. *N,N*-di(2,3-epoxypropyl)aniline (DEA) and 4-(2',3'-epoxypropoxy)benzophenone (EBP) were prepared from epichlorohydrin of aniline and 4-hydroxybenzophenone, respectively.⁶ The epoxy prepolymer was a low molecular weight liquid diglycidyl ether of bisphenol-A (DGEBA) (E-51 by Shanghai Resin Factory). The hardener was reagent grade 4,4'-diaminodiphenylmethane (DDM) (Shanghai No. 1 Chemical Reagent Factory).

PHOTOPOLYMERIZATION OF ELASTOMER

The photopolymerization in organic solvent (benzene for ETPnBA, dioxane for CTPnBA) was carried out in a 10 mL Pyrex polymerization tube placed in a water bath at 40 ± 0.1°C. A 300 W high pressure mercury lamp served as the radiation source; a CuSO₄ solution filter was used to cut off the light under 365 nm wavelength for photopolymerization of ETPnBA, no filter being used for CTPnBA. Before photopolymerization, the solution was bubbled with purified nitrogen to remove the oxygen dissolved in the system. After polymerization was over, the polymer was precipitated with methanol.

EPOXY RESIN CURING

The curing of epoxy resin was carried out by a two-step curing process. In the first step, the medium molecular weight rubber was precooked with the epoxy resin and DDM by heating in acetone for 30 min so as to complete the prereaction. Then the solvent was removed gradually, and the mixture was degassed in a vacuum oven at about 85°C for about 20 min. In the second step, the resins were poured into plates approximately 55 × 25 mm and cured in a controlled oven at 90°C for 3 h and 180°C for 1 h. After cooling to room temperature naturally, the various samples for the mechanical tests were machined from these cured epoxy plates.

MEASUREMENTS

In the polymer analytical procedures, the elastomers were separated from the excess initiator, photosensitizer, and monomers by dissolving in acetone and reprecipitated with methanol for three to four times, and then dried at 80°C under vacuum for 24 h. Their molecular weights and molecular weight distributions of the purified elastomers were measured by GPC (Shimadzu LC-3A Model), in chloroform at 25°C, polystyrene as calibration standards. The viscosity average molecular weights of the elastomers were determined by an Ubbelohde viscometer in acetone at 25°C.⁷ The ultraviolet absorption spectra were measured in chloroform with a Shimadzu UV-240 ultraviolet spectrophotometer. The epoxy functionalities of ETPnBA and ETPnBA/GMA elastomers were determined by nonaqueous titration and ultraviolet spectra.^{8,9} The measurements of carboxyl functionality of CTPnBA and CTPnBA/AA elastomers were based on acid-base titration in organic solution as described in the literature.¹⁰

Tensile fracture and elongation of epoxy resins were measured at a crosshead speed of 100 or 5 mm/

Table I Functionality of nBA Polymers in Various Polymerization Conditions^a

No.	Temp (°C)	[nBA] (mol/L)	[DEA] × 10 ³ (mol/L)	[BP] × 10 ³ (mol/L)	<i>M_n</i> × 10 ⁻⁴	<i>f_{uv}</i> ^b	<i>f_t</i> ^c
1	40.0	1.77	8.62	8.60	2.6	1.8	1.6
2	40.0	2.09	46.2	8.60	3.0	1.8	2.1
3	40.0	2.09	8.62	1.50	4.0	0.9	0.9
4	20.0	2.09	8.62	8.60	3.6	1.6	1.6

^a Benzene as solvent.

^b *f_{uv}* = functionality measured by ultraviolet spectra.

^c *f_t* = functionality measured by nonaqueous titration.

min with Instron 1121 instrument. Tensile strength and Young's modulus were obtained simultaneously by this test. The tensile impact energy was calculated from the area under the stress-strain curve at high crosshead speed. Impact strength of these modified samples was measured by the Charpy impact test, each sample tested five to 10 times for obtaining an average value. The morphologies of impact fracture surfaces of samples were observed using scanning electron microscopy (SEM) with a magnification capability up to 20,000 \times . The fracture surfaces were coated with a layer of gold about 200 Å thick.

RESULTS AND DISCUSSION

Photosynthesis of Poly(*n*-Butylacrylate) Elastomers Containing Polyfunctional Epoxy Groups

Aromatic carbonyl compounds have been found to undergo very efficient photoreduction reactions in the presence of tertiary aliphatic amines producing aromatic ketyl radicals and radicals on aliphatic carbon centers α to the amine functional group.⁹ However, the experimental results show that the functionality of polymers formed was surprisingly low (see Table I). They apparently indicate that, on the average, one end epoxy group per chain could be found in the case of photopolymerization of *n*-butylacrylate initiated with the BP-DEA system. This may be attributed to the deactivation of the active chain by the semipinacol radicals formed from BP via electron transfer between BP and DEA.⁹

To attach two epoxy terminal groups on the chain of poly(*n*-butylacrylate), 4-(2,3-epoxypropyl)benzophenone (EBP) instead of BP was used

Table II Functionality of GMA/*n*BA Copolymers Initiated by DEA-EBP^a

No.	$V_{\text{GMA}}/V_{\text{nBA}}$	$M_n \times 10^{-4}$	f_m^b	f_p^c
E1	0	3.60	2.9 (f_e^d)	0
E2	0.01	3.65	6.9	4
E3	0.02	3.70	10.9	8
E4	0.04	3.80	19.0	16.1
E5	0.06	3.74	27.0	24.1

^a [DEA] = 9.5×10^{-3} mol/L, [EBP] = 7.5×10^{-3} mol/L, benzene as solvent.

^b f_m = functionality data by measurement.

^c f_p = functionality of pendent group.

^d f_e = functionality of end group ($f_m = f_p + f_e$).

Table III Molecular Weight and Functionality (f) of *n*BA Polymers with Various Concentration of DTPA^a

No.	[DTPA] (mol/L)	d^b	$M_n^c \times 10^{-5}$	$M_n \times 10^{-4}$	f
1	0.13	4.4	1.56	3.54	1.6
2	0.10	5.0	1.79	3.58	2.0
3	0.08	5.0	1.83	3.59	2.3
4	0.056	5.5	1.98	3.60	1.7

^a Dioxane as solvent.

^b d = molecular weight dispersion measured by GPC.

^c M_n = viscosity-average molecular weight.

as the photosensitizer to initiate the polymerization of *n*BA. As expected, the polymeric chain of sample 1 (E1) listed in Table II possesses two terminal epoxy groups, $f_e = 2.9$. The detailed kinetic study on the photopolymerization of *n*BA initiated by DEA and EBP has been discussed in another paper.⁶ The reactive scheme proposed for the photopolymerization of *n*BA may occur as follows: The absorption of light quanta by EBP is the first step, resulting in excitation of EBP to excited state, the secondary step is one where the excited state EBP* forms an exciplex with the DEA. Photoproduction of free radicals from DEA and EBP* via the electron-transfer mechanism follows. DEA free radical intermediate was shown to initiate the polymerization of *n*BA; the semipinacol radical formed from EBP is reactive to terminate the active chain of poly(*n*-butylacrylate):

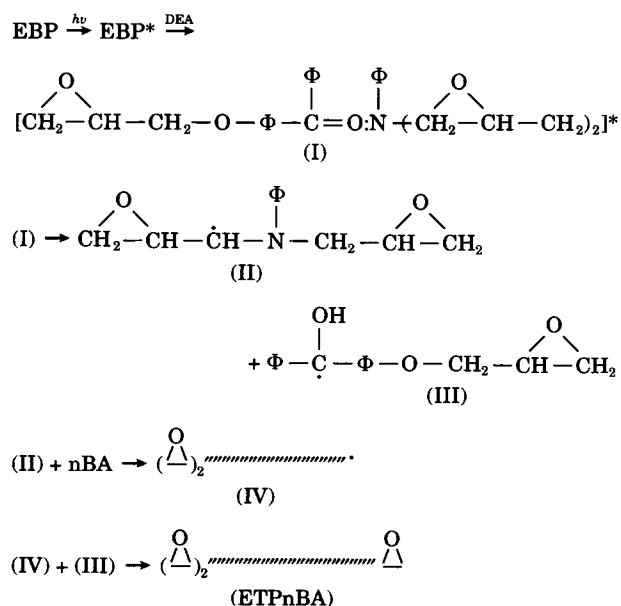


Table IV Functionality of AA/nBA Copolymer Initiated with DTPA^a

No.	V_{AA}/V_{nBA}	$M_n \times 10^{-4}$	f_m^b	f_p^c
C1	0	3.60	1.8 (f_e^d)	0
C2	0.01	3.62	6.8	5
C3	0.02	3.59	10.7	8.9
C4	0.04	3.61	20.8	19
C5	0.06	3.57	28.7	26.9

^a [DTPA] = 0.13 mol/L, dioxane as solvent.^b f_m = functionality data by measurement.^c f_p = functionality of pendent group.^d f_e = functionality of end group.

In order to increase interaction between elastomer chain and epoxy matrix, the copolymerization of nBA and glycidylmethacrylate (GMA) was studied in solution at 40°C using the EBP-DEA photoinitiation system. As seen in Table II, the functionality of ETPnBA increases with the increase of the volumetric ratio of GMA/nBA.

PHOTOSYNTHESIS OF POLY(*n*-BUTYLACRYLATE) ELASTOMERS CONTAINING POLYFUNCTIONAL CARBOXYL GROUPS

For comparison, the carboxyl terminated poly(*n*-butylacrylate) elastomer was synthesized in dioxane solution of nBA at 40°C using dithiodipropionic acid (DTPA) as photoinitiator under the irradiation of

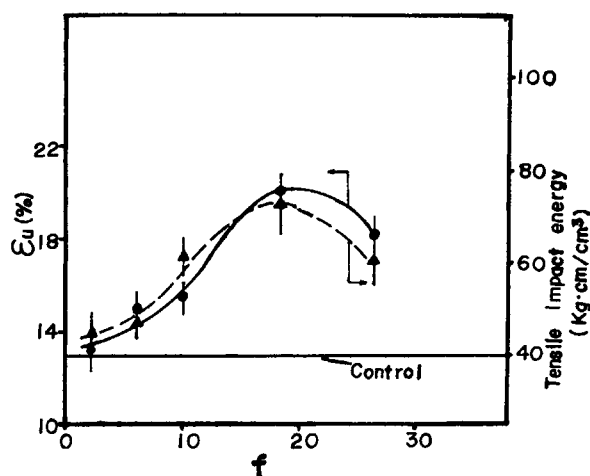


Figure 1 Tensile impact energies and ultimate elongation of ETPnBA-modified epoxy resin vs. functionality of rubber. Crosshead speed: 100 mm/min, ER/DDM/ETPnBA = 100/25/5 (pbw).

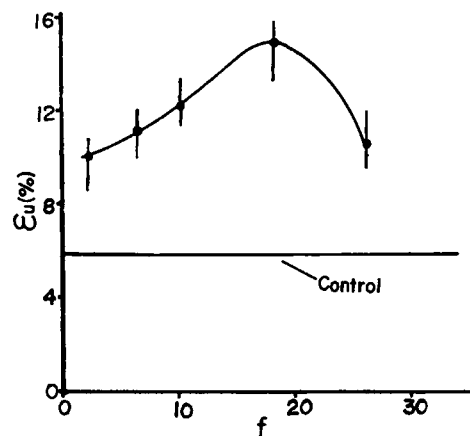


Figure 2 Ultimate elongation of ETPnBA-modified epoxy resin vs. functionality of rubber. Crosshead speed: 5 mm/min, ER/DDM/ETPnBA = 100/25/5 (pbw).

above 300 nm. It has been suggested that symmetrical difunctional chain transfer agents be used for synthesis of telechelic polymers.¹¹ DTPA was selected in the present work as photoinitiator to introduce the terminal functional groups onto the chain of poly(*n*-butylacrylate). The solution photopolymerization of nBA in dioxane was investigated with respect to the concentration of DTPA. The reaction in the DTPA molar concentration from 0.056 to 0.13 shows a gradual decrease of molecular weight (M_n , M_w) and degree of molecular weight dispersion (d), while the functionality of polymers is around $f = 2$ (Table III). It appears that carboxyl groups are end-blocked; consequently, DTPA behaves as either photoinitiator or chain transfer agent. For the same reason as ETPnBA, acrylic acid was added to copolymerize with nBA in dioxane initiated by DTPA. As shown in Table IV, a set of carboxyl terminated polyfunctional poly(*n*-butylacrylate) (CTPnBA) was obtained, the functionality of carboxyl groups in the copolymers was 6.8–28.7 in a range of 0.01–0.06% (vol %) of the acrylic acid in the monomer mixture.

MECHANICAL PROPERTIES

The effect of rubber functionality on the mechanical properties of DGEBA/rubber blends was studied with ETPnBA and CTPnBA. The tensile impact energies and elongation of DGEBA modified with a series of epoxy-terminated copoly(*n*-butylacrylate and glycidylmethacrylate) elastomers (ETPnBA) is shown in Figures 1 and 2. The results indicate that the impact energy and elongation increase with

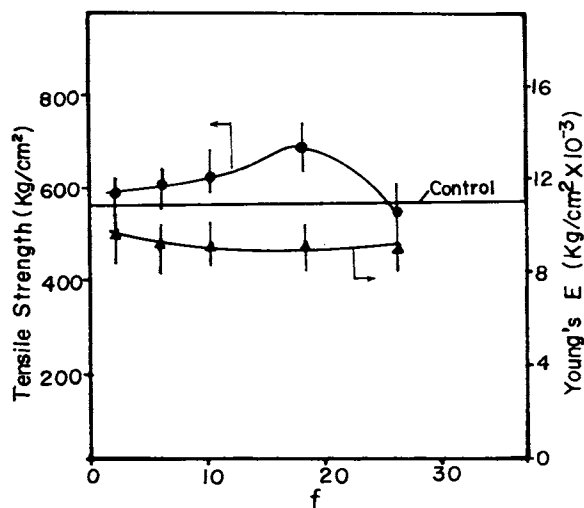


Figure 3 Tensile strength and Young's modulus of ETPnBA-modified epoxy resin vs. functionality of rubber. Crosshead speed: 5 mm/min, ER/DDM/ETPnBA = 100/25/5 (pbw).

increasing of epoxy group content of the elastomers, but show a maximum value in the region where the epoxy functionality of the elastomer (ETPnBA) is about 19. The toughest sample had a tensile impact energy 85% (fast crosshead speed) and a elongation 150% (slow crosshead speed) greater than that of control. The results presented in Figures 1 and 2 show that there exists an optimum functionality of elastomer for the toughening of DGEBA modified with ETPnBA.

The tensile strength and Young's modulus of DGEBA/ETPnBA systems, as shown in Figure 3, indicate that the tensile strength of DGEBA modified with ETPnBA has a small increase with respect to that of control. However, Young's modulus of modified epoxy resins decreases slightly with the increase of functionality of ETPnBA. This behavior is expected since the ETPnBA rubbers with medium molecular weights, M_n up to 3.7×10^4 , are in general higher than that of RLPs (reactive liquid polymers), in which the molecular weight were usually in the range of $3.0-6.0 \times 10^3$.¹²

The effects of the rubber content on the mechanical properties of DGEBA/ETPnBA blends are shown in Figures 4 and 5. Initially, the elongation increases with increasing elastomer content, but only improves slightly when the content of ETPnBA is over 5% (pbw). In addition, the tensile strength and Young's modulus decrease sharply with increasing elastomer content. Therefore, to increase the fracture toughness of the cured epoxy resin without a significant decrease of other mechanical

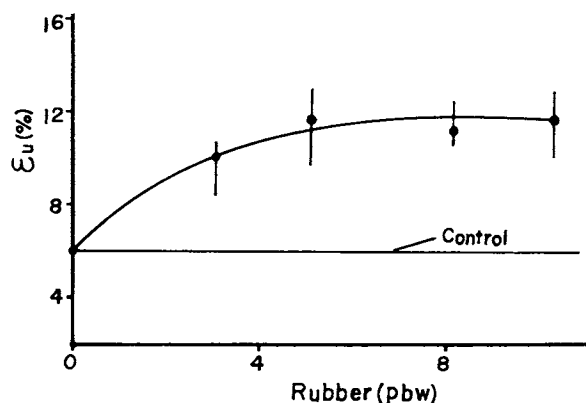


Figure 4 Ultimate elongation of ETPnBA(E3)-modified epoxy resin with various rubber content. Crosshead speed: 5 mm/min, ER/DDM = 100/25 (pbw).

properties, it appears that the optimum content of the rubber in DGEBA/ETPnBA blend should be around 5% (pbw). This is one of the advantages to use of the novel higher functionality elastomer with medium molecular weight in the toughening of epoxy resin. The amount of the elastomer used in this system was much less than that of low molecular weight RLP, which is generally in the range of 10–20% (pbw) as reported previously.¹³ This means the addition of smaller amount of ETPnBA elastomer results in significant improvement in the toughness of DGEBA system.

The toughening effect of CTPnBA/DGEBA system is found to be a different behavior from ETPnBA/DGEBA system. The elongation, tensile

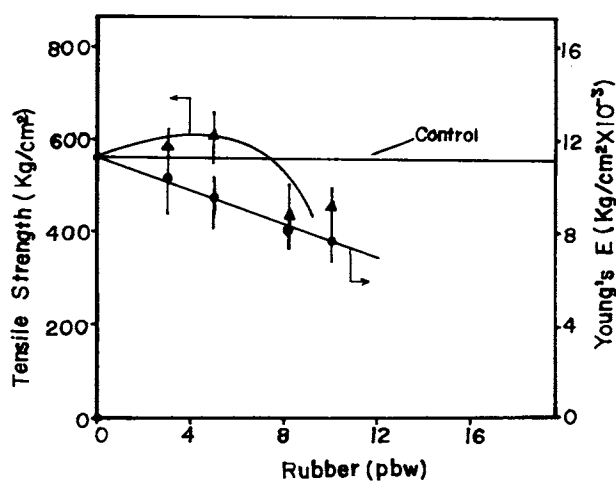


Figure 5 Tensile strength and Young's modulus of ETPnBA(E3)-modified epoxy resin with various rubber contents. Crosshead speed: 5 mm/min, ER/DDM = 100/25 (pbw).

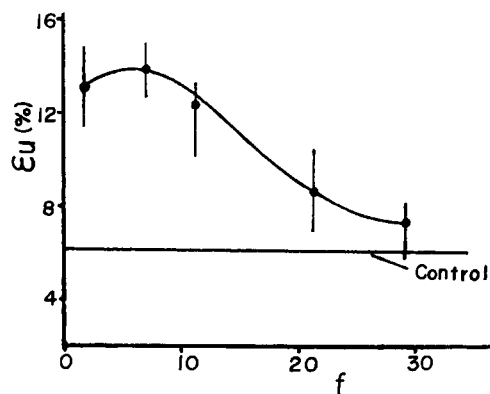


Figure 6 Ultimate elongation of CTPnBA-modified epoxy resin vs. functionality of rubber. Crosshead speed: 5 mm/min, ER/DDM/CTPnBA = 100/25/5 (pbw).

strength, and Young's modulus of DGEBA modified with a series of carboxyl-terminated copoly(*n*-butyl acrylate and acrylic acid) elastomers (CTPnBA) are shown in Figures 6 and 7 respectively. These results indicate that the elongation of DGEBA modified with CTPnBA elastomers decreases with the increasing of functionality of carboxyl group. The same is true for the tensile strength and Young's modulus.

The effect of the functionality of CTPnBA on the impact strength of modified DGEBA system is given in Table V. The result reveals that the impact strengths of epoxy resins are slightly increased by incorporating the carboxyl-terminated elastomers. But with increasing levels of the pendant carboxyl group on the backbone of elastomer, the impact strengths of modified epoxy resins decrease. This is consistent with above tensile test results.

The observation could be explained by the difference in the degree of compatibility of elastomers in the DGEBA and the morphology of the ETPnBA and CTPnBA in the matrix. A detailed discussion will follow in the morphology observation.

Morphologies

The effect of the reactive group and functionality of the elastomers on morphology of phase separation was studied for blending with 5% elastomer. SEM examination of the fracture surface of ETPnBA/DGEBA specimens is shown in Figure 8. Figure 8(a) is an SEM view of the unmodified epoxy specimen, in the fast fracture region, the surface is smooth which indicates much less yielding of the material. At low functionality, $f = 6$ –19, as shown in Figures 8(c)–8(e), there is localized shear yielding that ap-

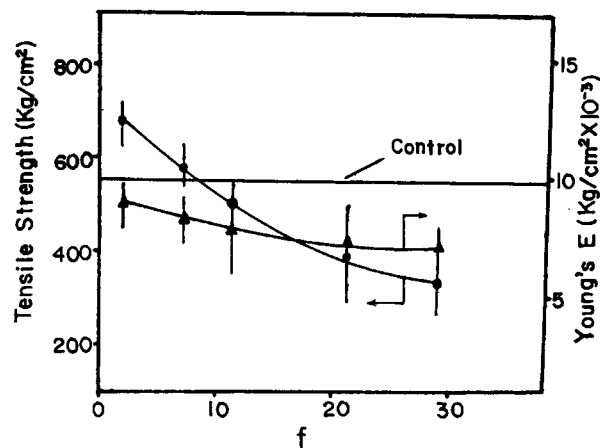


Figure 7 Tensile strength and Young's modulus of CTPnBA-modified epoxy resin vs. functionality of rubber. Crosshead speed: 5 mm/min, ER/DDM/CTPnBA = 100/25/5 (pbw).

pears to be associated with holes and spherical domains of the elastomer. The presence of dispersed phase could be responsible for the toughening action. With increasing functionality of the elastomers, there is a gradual decrease of the size of the dispersed phase, in addition to particles of a few micrometers, the rubber is also present as a finely dispersed phase of particles having diameters of the order of a few tenths of a micrometer. However, when the functionality is higher than 19 [Fig. 8(f)], most of the dispersed phase is found to be in particles of a few tenths of a micrometer in diameter. The results presented in Figure 8 suggest that there exists a multiple distribution of particle sizes in the ETPnBA elastomer modified epoxy resins. It is well known that, for effective toughening of the epoxy resin, the rubbery elements have to be miscible and grafted, to a certain degree, to the DGEBA matrix. However, an excessive compatibility and an excessive amount of chemical interaction between the DGEBA and the rubber modifier might lead to the formation of a

Table V Effect of Functionality (f) of CTPnBA on Impact Strength (IS)

No.	f	IS (kg cm/cm ²)
0	—	6.7 (±1.1)
C1	1.8	8.4 (±2.0)
C2	6.8	8.0 (±4.2)
C3	10.7	7.4 (±2.5)
C4	20.8	6.8 (±3.3)
C5	28.7	7.1 (±0.9)

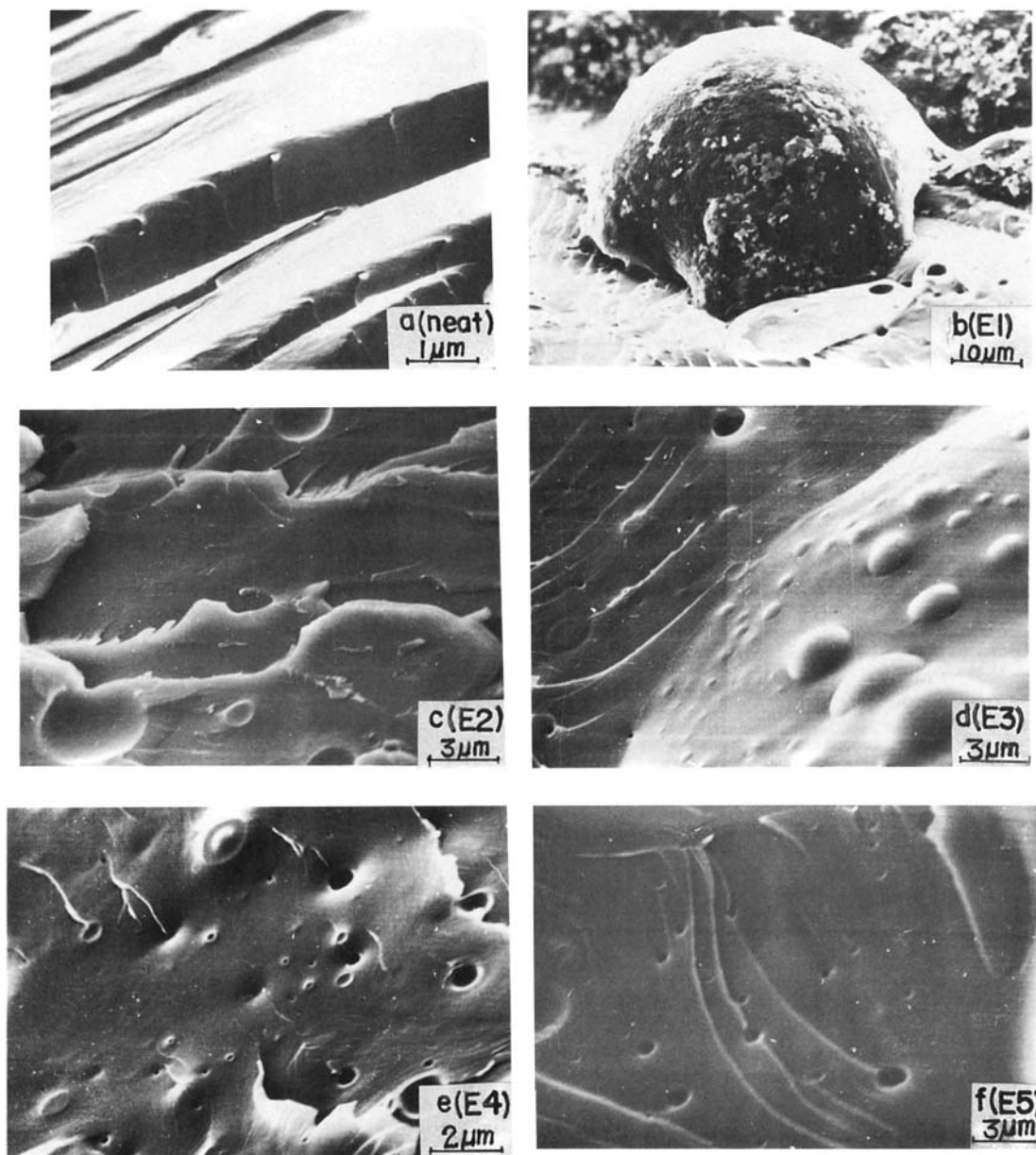


Figure 8 Effect of functionality of ETPnBA rubber on morphology of fracture surfaces of ETPnBA-modified epoxy resin. ER/DDM/ETPnBA = 100/25/5 (pbw).

single phase which is an undesirable morphology. Clearly, the effect of the functionality of ETPnBA on the morphology of ETPnBA/epoxy blends is consistent with its influence on the mechanical properties of this system.

The morphology of the fracture surfaces of epoxy resin modified with CTPnBA elastomers reveal a general decrease in diameter of elastomeric particles with the increasing of the functionality of CTPnBA, as shown in Figure 9. Meanwhile, owing to the clustering of elastomeric particles, the elastomer is

present in the aggregates as a dispersed phase having diameters of the order of 15–30 μm . In addition, the size of the aggregates increases with the increasing of the functionality of CTPnBA. Associated with the lower toughness of DGEBA/CTPnBA blends, it appears that the toughening effect of CTPnBA is not as evident as that of ETPnBA in the modified epoxy resin system; this could be attributed to aggregation of its elastomeric particles.

However, in the case where the elastomers contain only a terminated functional group, there is no

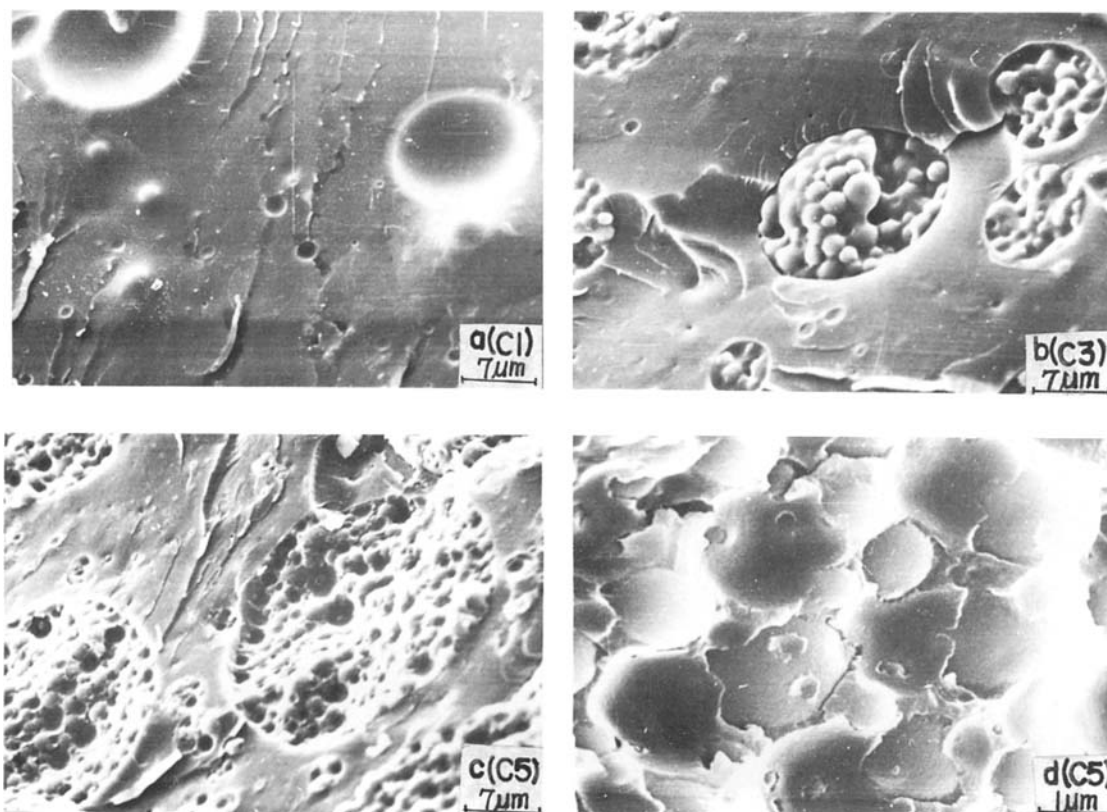


Figure 9 Effect of functionality of CTPnBA rubber on morphology of fracture surfaces of CTPnBA-modified epoxy resin. ER/DDM/CTPnBA = 100/25/5 (pbw).

significant difference in their morphology between ETPnBA- and CTPnBA-modified DGEBA systems. This behavior indicates that the pendent carboxyl groups of the elastomeric backbone would not be beneficial to the toughening of DGEBA/CTPnBA blend. The combination of high functionality and high molecular weight of CTPnBA might be responsible for the aggregation of the CTPnBA particles.

At high magnification, by carefully scrutinizing the dispersed phase of the composition, the rubbery particles display a core-shell structure [Fig. 9(d)]. Romanchick showed that the core-shell structure was a result of the rubber being depleted by the epoxy resin matrix to form domains in which the interior of the domain was richer in epoxy resin and the shell was richer in elastomer,¹⁴ although Hemi has said that the rubber occupies mainly the core.¹⁵ In our case, the shell is mainly composed of the elastomer.

CONCLUSIONS

A set of novel ETPnBA and CTPnBA elastomers with medium molecular weight up to 3.8×10^4 (M_n)

and functionality in the range of 2–29 was obtained by photopolymerization for use as modifiers in epoxy resin toughening. The mechanical properties and morphology of ETPnBA/DGEBA blends and CTPnBA/DGEBA blends with various degrees of elastomeric functionality cured with DDM have been studied. The morphology of these blends is strongly influenced by the kind of functional group, its site on the chain, and functionality of the rubbery modifiers, displaying very different mechanical behavior.

It has shown that the presence of a multiple distribution of the dispersed phase is a necessary condition to improve the toughness of epoxy resin modified with the rubber. From the results in the present work, ETPnBA with functionality of $f = 19$ is a suitable component for this purpose. Thus, the pendant epoxy groups of ETPnBA, to a certain extent, display an important role for the toughening of ETPnBA/DGEBA blends. On the contrary, in the case of CTPnBA, the pendent carboxyl groups cause the aggregation of rubbery particles in the epoxy matrix, which might be responsible for poor mechanical properties of CTPnBA/DGEBA blends. Finally, use of a smaller amount of the higher functionality ETPnBA with medium molecular weight

provides effective toughening; thus, compared with the low molecular weight RLP, the potential of ETPnBA as a modifier for epoxy resins is promising.

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