

Matrix Viscoelasticity: Controlling Factor in the Rubber Toughening of Epoxy Resins

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Synopsis

Our limited success in toughening methylene dianiline (MDA)-cured Epon 828, using varying rubber types, led to a study of the role of the matrix viscoelasticity in the toughening process. Two rubber types, with different interfacial bonding capabilities, poly(*n*-butyl acrylate)/15 wt % acrylonitrile/2 wt % acrylic acid and poly(*n*-butylacrylate)/15 wt % acrylonitrile, were incorporated into systems containing varying amine concentrations to control crosslink density. Impact strengths of controls and rubber-modified compositions increased with excess amine concentrations up to 70%. The impact strengths for the poly(*n*-butyl acrylate)/15 wt % acrylonitrile/2 wt % acrylic acid rubber-modified compositions were greater than their equivalent controls, with the effect being greater at a lower crosslink density. This study confirmed that the matrix viscoelasticity is the controlling parameter in the toughening process. The degree of rubber-epoxy interfacial bonding is also an important parameter to consider, if the matrix viscoelasticity permits toughening. A modified stress response model was used to explain the toughening phenomenon.

INTRODUCTION

In a previous study¹ the compatibility of poly(*n*-butyl acrylate) rubber and its effect on the epoxy composite mechanical properties were studied, using stoichiometric equivalence of amine to epoxy, for the methylene dianiline (MDA)-cured system. The results showed that PnBA rubber-epoxy compatibility could be improved by the addition of acrylonitrile (AN) and acrylic acid (AA) as co- and terpolymers. The addition of the acrylonitrile raises the solubility parameter of the rubber (due to the high polarity of AN), while the addition of AA also increases the number of bonding sites on the rubber molecules, causing an improvement in the interfacial adhesion between rubber and epoxy. A change in compatibility effected a subsequent change in the precipitated properties of the rubbery second phase. Improved compatibility initially resulted in smaller, more uniformly sized particles. At high compatibility, the rubber became trapped in solution and less rubber precipitated.

The precipitation study lead to an investigation dealing with the effects of varying compatibility and/or interfacial bonding on the mechanical properties of the rubber-modified compositions. The results showed that for a poorly compatible, weakly bonded, two-phase system, composite material properties are degenerated as compared to those of the equivalent control. The presence of the rubber had a detrimental effect on the impact strength

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of the composite. As compatibility and/or interfacial bonding improved, the impact strength initially improved but leveled off at a value which was near to the strength of the control. For this relatively high T_g MDA-Epon 828 system, the rubber did not produce any significant improvement in impact strength. It was concluded that the rubber is an important parameter in the toughening mechanism, but its importance was likely secondary to that of the role of the matrix viscoelasticity, which is the subject of the present work.

For this study two rubber types, PnBA/15% PAN/2% PAA and PnBA/15% PAN were used throughout. It was felt that the former rubber represents an initially compatible rubber, which after precipitation would be bonded, but not too tightly, to the matrix. The variable parameter was the concentration of the curing agent. By increasing the concentration of MDA (referred to as % excess amine) the crosslink density of the epoxy matrix could be effectively reduced, and the flexibility of the network chains increased. The ability of the rubber to induce greater toughening in a more flexible network would suggest the dominance of matrix viscoelasticity in the toughening mechanism.

EXPERIMENTAL

Copolymerization Method and Characterization of Rubber

The procedure for the co- and terpolymerization of the PnBA/PAN, PnBA/PAA, and PnBA/PAN/PAA rubbers is similar to that for the PnBA rubber.² Acrylonitrile (0–42% by weight) and acrylic acid (0–20% by weight) were polymerized together and separately with butylacrylate using 4,4-azobis(4-cyanovaleric acid) and dithiodiglycolic acid as the initiator and chain transfer agent, respectively. The reaction proceeded by bulk polymerization under continuous mixing in a reaction vessel purged with nitrogen gas. The polymerization temperature was maintained at 72°C because of acrylonitrile's relatively low boiling point (77°C). After polymerization the rubbers were washed with an acetone and water separation technique to remove all unreacted materials. Molecular weight fractions were obtained by slowly varying the acetone to water ratio during the wash cycle.

Number (\bar{M}_n) and weight (\bar{M}_w) average molecular weights of the rubbers were measured using GPC. The GPC was calibrated using low polydispersity PnBA/PAN/PAA rubber fractions. Actual number average molecular weights for the weight fractions were measured using a Knauer Vapor Pressure Osmometer. Rubber functionality was measured by titration with potassium hydroxide.²

Preparation of High Speed Tensile Test Samples

Plates for the high speed tensile test samples were made using a two-step cure cycle. Ten weight parts Epon 828 with reaction catalyst (1% *tetra-n*-butylammonium iodide) were mixed with 1 weight part rubber and placed in an oven for 2 h at 120°C. This cure step insures the formation of the epoxy-rubber adduct prior to the final cure.

In the second step of the curing cycle, melted (approx. 100°C) 4,4-methylenedianiline (MDA), from stoichiometric equivalent to 100% excess amine, was added to the epoxy rubber mixture. After hand stirring, the solution was cast between two glass plates sprayed with Miller-Stephenson MS-136 Hot Mold Release agent and placed in the oven at 120°C for 1 h. The temperature was raised to 150°C and the samples were post cured for 2 h before slowly cooling to room temperature. Mechanical test specimens were machined from the plates.

Impact strengths were measured using a high speed Tensile Impact Tester previously described.³ Ultimate stress, ultimate strain, modulus, energy to break, and speed of break were calculated for all samples. Six to twelve samples from each group were broken. Average values, along with the standard deviations, are reported.

Impact strength, were also measured by the standard Notched Izod test (Standard ASTM D256-56) using the Testing Machines Inc. (TMI) Model 52004.

An Instron Table Model TM-S was used for the low strain rate tensile testing. Analysis procedures followed the ASTM D638-68 standard. The crosshead speed was 0.2 in./min.

RESULTS

Toughening at High Strain Rates (High Speed Tensile Impact Test)

Energy to break for PnBA/15% AN/2% AA rubber modified compositions as a function of amine concentration is presented in Figure 1. An increase in amine concentration results in an increase (up to 70% excess amine) in energy to break. The increased flexibility of the network chains allows better dissipation and absorption of the applied strain energy. The rubber-modified compositions in general had higher energies to break than their

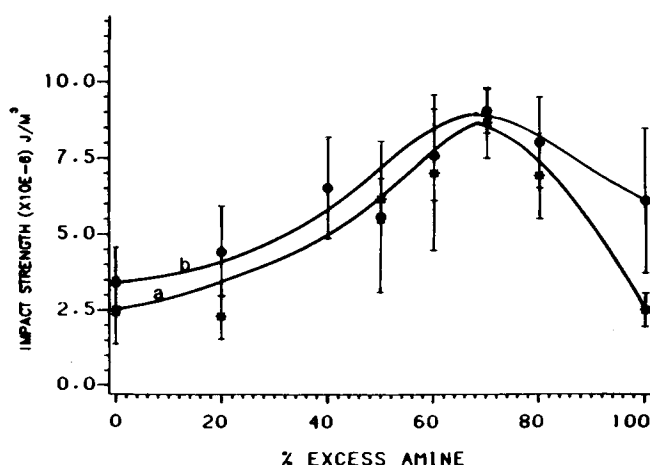


Fig. 1. Tensile impact test, energy to break vs. % excess amine: (a) control and (b) PnBA/15% AN/2% AA rubber-modified.

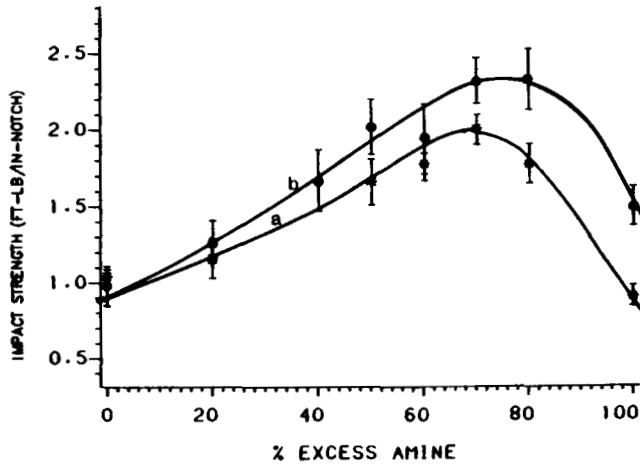


Fig. 2. Izod impact strength vs. % excess amine: (a) control and (b) PnBA/15% PAN/2% PAA rubber-modified.

equivalent control. At amine concentrations of 80% excess and above, the impact strengths of the controls drop off more sharply as compared to those of the rubber-modified compositions. In the controls, a point is reached where the network becomes too flexible, crosslinks are spaced too far apart, and network strength drops. In the rubber-modified compositions at the higher amine concentrations, the network also becomes flexible, but the rubber helps maintain the overall strength of the epoxy network, and energy to break does not drop off as drastically.

Energy to break, measured with the Izod impact tester at varying amine concentrations (Fig. 2), gives results comparable to the high speed tensile impact-measured energy to break. Rubber-modified compositions are somewhat tougher than the controls. The degree of toughness improvement is greater at the higher amine concentrations.

Elongation to break and ultimate stress curves from the high speed experiments are presented in Figures 3 and 4, respectively. Increasing amine

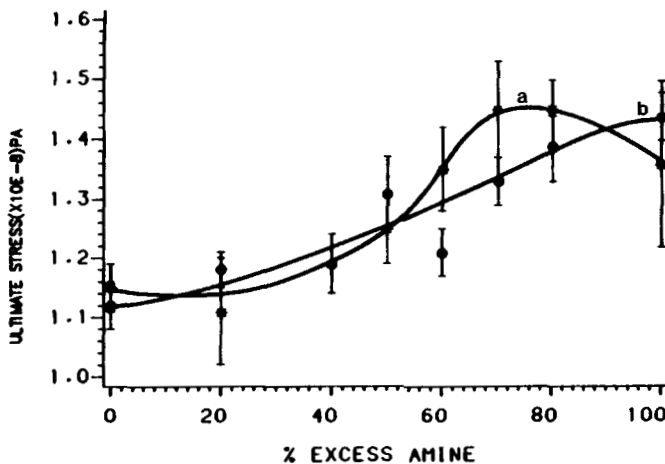


Fig. 3. Tensile impact test, ultimate stress vs. % excess amine: (a) control and (b) PnBA/15% PAN/2% PAA rubber-modified.

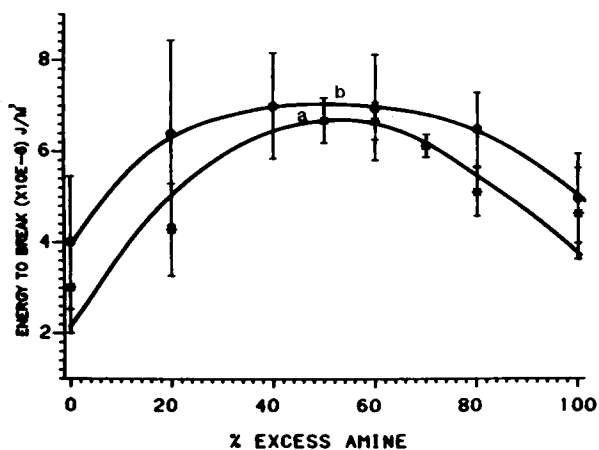


Fig. 4. Tensile impact test, elongation to break vs. % excess amine: (a) control and (b) PnBA/15% PAN/2% PAA rubber-modified.

concentration gives a slight increase in ultimate stress for both control and rubber-modified compositions. Elongation to break, as with the energy to break, peaks around 70% excess amine. The rubber-modified compositions show greater values for elongation to break than the equivalent controls.

Modulus remains relatively constant with varying amine concentrations (Fig. 5). Values of moduli for rubber compositions were less than the controls.

Toughening at Slow Strain Rates (Instron)

Energy to break, ultimate stress, elongation to break, and modulus for PnBA/15% AN/2% AA compositions, with varying amine concentrations, tested at slow strain rates show the same trends as samples fractured at the high strain rates. Energy to break increased with increasing amine concentrations to 60% excess amine (Fig. 6), representing a shift of about 10% from the peak in the high speed tensile energy to break curve. Overall, strengths measured with the Instron were not significantly different from those measured at high strain rates.

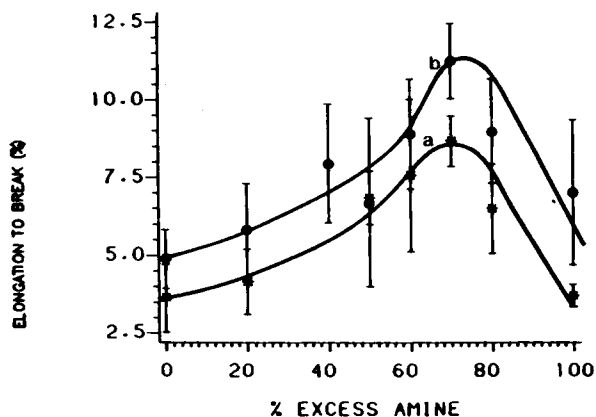


Fig. 5. Tensile impact test, modulus vs. % excess amine: (a) control and (b) PnBA/15% PAN/2% PAA rubber-modified.

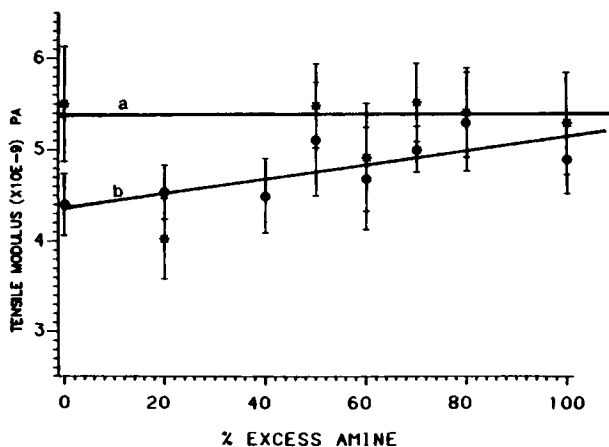


Fig. 6. Instron, energy to break vs. % excess amine: (a) control and (b) PnBA/15% PAN/2% PAA rubber-modified.

Matrix Viscoelasticity Study Using PnBA/15% PAN Rubber

A second rubber type, PnBA/15% PAN, which exhibits less matrix bonding, was incorporated into MDA-cured epoxy compositions at varying amine concentrations. The objective of this study was to determine whether poorly bonded rubber would result in less toughening at the higher amine concentrations. Results are presented in Figures 7-10. There appears to be no significant difference in material properties of epoxies modified with either the PnBA/15% PAN/2% PAA or PnBA/15% PAN rubbers up to amine concentrations of 50% excess. At the higher amine concentrations, as a result of poor rubber-epoxy interfacial bonding, the strength of the PnBA/15% PAN rubber-modified compositions is lower than either the better bonded PnBA/15% PAN/2% PAA-modified or control formulations. This confirms that the degree of rubber-epoxy interfacial bonding is an important parameter to consider if the matrix viscoelasticity permits toughening.

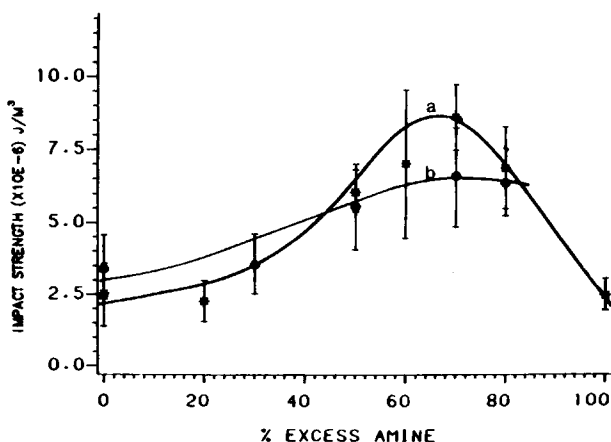


Fig. 7. Tensile impact test, energy to break vs. % excess amine: (a) control and (b) PnBA/15% PAN rubber-modified.

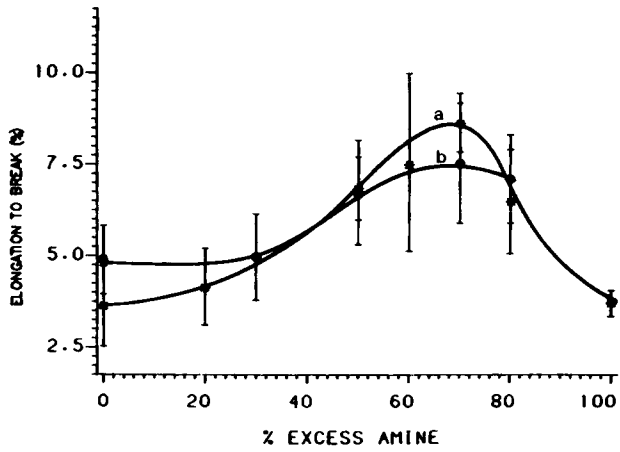


Fig. 8. Tensile impact test, elongation to break vs. % excess amine: (a) control and (b) PnBA/15% PAN rubber-modified.

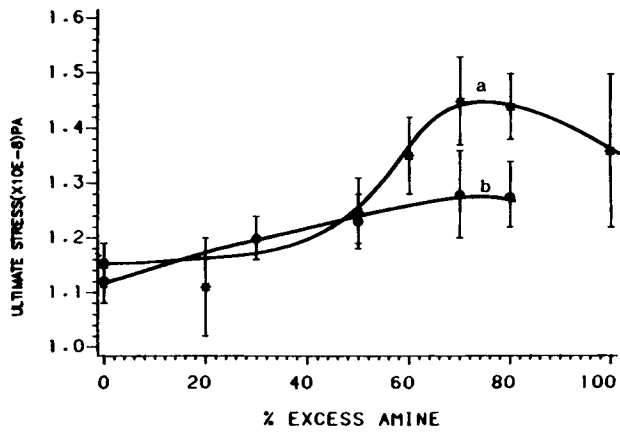


Fig. 9. Tensile impact test, ultimate stress vs. % excess amine: (a) control and (b) PnBA/15% PAN rubber-modified.

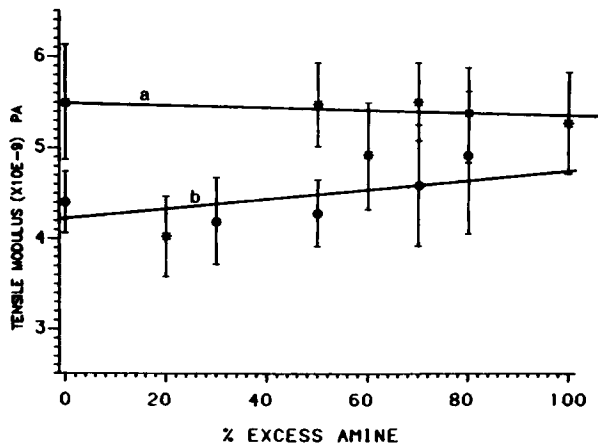


Fig. 10. Tensile impact test, modulus vs. % excess amine: (a) control and (b) PnBA/15% PAN rubber-modified.

If the matrix is stiff, and no plastic deformation occurs, then impact strengths of the composites containing bonded and nonbonded rubber are approximately the same.

Comparison of the Mode of Failure for Samples Broken at Low (With Instron) and High (With Tensile Impact) Strain Rates

Interesting insights about the mechanism of rubber-modified epoxy failure can be obtained from the examination of the fractured specimens and correlation of the observations with the recorded stress-strain curves. Even without extensive fracture mechanics or scanning electron microscopy studies, examination of the broken samples gives valuable information about the failure mechanism.

The control samples, of close to stoichiometric equivalence (one-phase system, 0–20% excess amine), fail by brittle failure at high strain rates and exhibit only slight amounts of plastic deformation at the low strain rate. No evidence of yielding is observed in either the Instron or tensile impact specimen. Observations of the fracture surface reveals a rough face with many "finger" markings (Fig. 11). The rubber-modified samples (0–20% excess amine) exhibit whitening when broken at the high strain rate. Samples broken at the low strain rate do not exhibit any obvious whitening. For these rubber-modified compositions at amine concentration near stoichiometric equivalence, whitening does not appear to increase the toughening behavior. Instron and tensile impact test stress-strain curves reveal no major yielding or plastic deformation in either the control or rubber-modified compositions.

At the intermediate amine concentration (40–50% excess amine), some interesting observations were made. Rubber-modified compositions fractured with the Instron exhibit yielding in the matrix. Many samples show the formation of shear bands (shear bands appear white) 45–50° to the fracture plane (Fig. 12). The same samples, broken at the high strain rate,

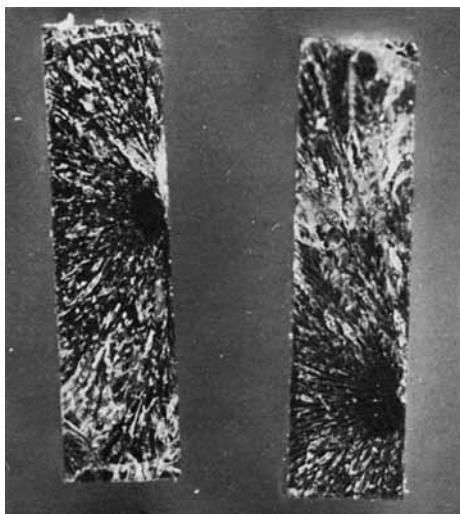


Fig. 11. Fracture surface of a MDA-cured (20% excess amine) control.

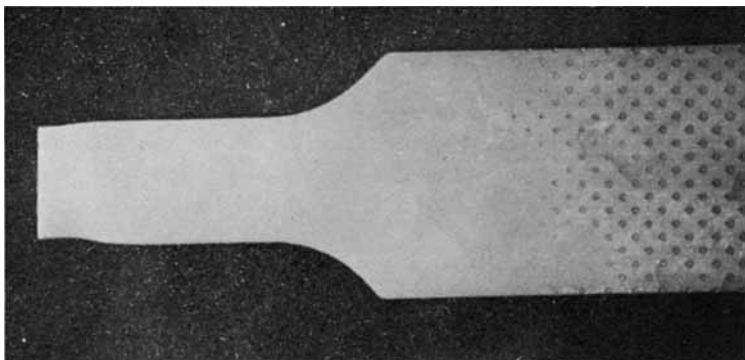


Fig. 12. Presence of a shear band in a PnBA/15% PAN/2% PAA rubber-modified MDA-cured (50% excess amine) epoxy.

whitened, with no evidence of shear banding (Fig. 13). Stress-strain curves for the 40% excess amine samples showed a leveling off in stress for samples broken with the Instron (no strain softening) and strain softening in the samples fractured with the tensile impact test. The whitening phenomenon appears to result in strain softening. Strain softening was also observed in the 50% and 60% excess amine rubber-modified compositions. Instron and tensile impact test stress-strain curves for the equivalent 50% and 60% excess amine controls showed strain softening. The Instron test specimen exhibited the formation of shear bands (clear) with some necking just prior to failure (Fig. 14). Tensile impact specimens did not exhibit any observable shear banding or necking.

At the highest amine concentrations (70–100% excess amine), the epoxy matrix is becoming ductile, and a greater degree of plastic deformation and yielding is observed in the control and rubber-modified compositions. The slow strain rate fractured rubber-modified specimens exhibit shear banding with evidence of necking, tearing (Fig. 15), and in some cases, “butterflies” are present (Fig. 16). Sternstein et al.⁴ have identified these patterns as regions of crazing in rubber-modified polystyrene systems. Gazit² has also observed butterfly patterns in a few rubber-modified DMHDA-cured Epon 828 epoxy systems and feels that this might represent the initiation of crazes by the rubber particles. To date, however, there exists no firm evidence that crazing occurs in high glass transition temperature rubber-modified

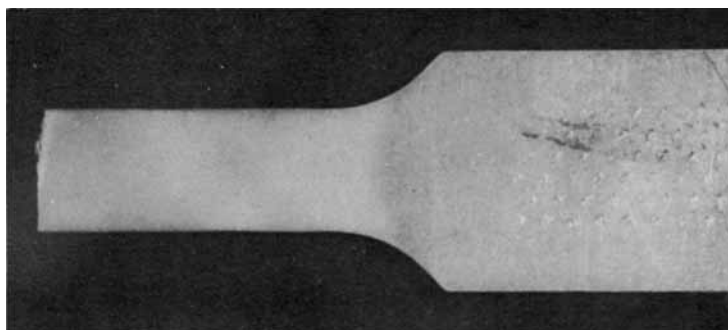


Fig. 13. Stress whitening in rubber-modified sample fractured at high strain rates.

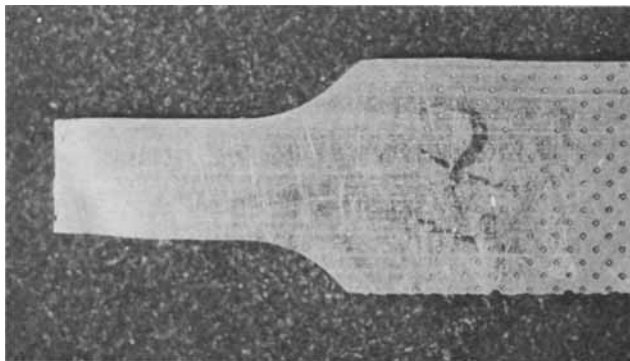


Fig. 14. Shear band, with evidence of necking, in a 60% excess amine epoxy control.



Fig. 15. Evidence of ductile behavior (tearing) in an 80% excess amine MDA-cured rubber-modified sample.

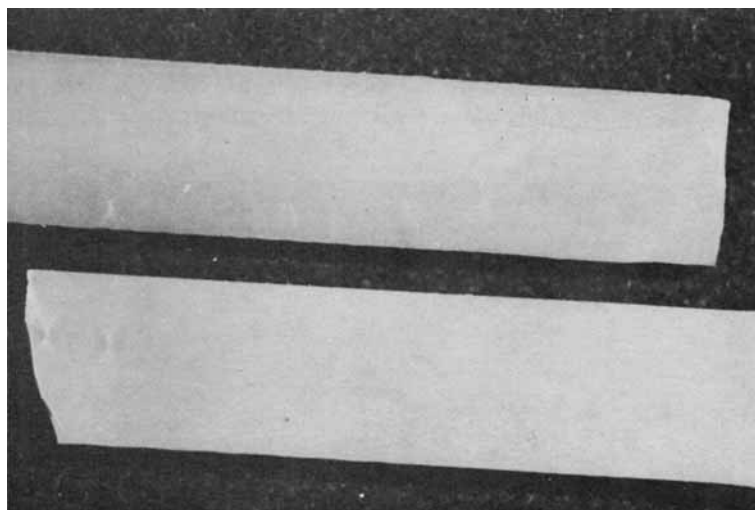


Fig. 16. Photograph showing "butterfly" patterns in a PnBA/15% PAN/2% PAA rubber-modified MDA-cured (70% excess amine) epoxy.

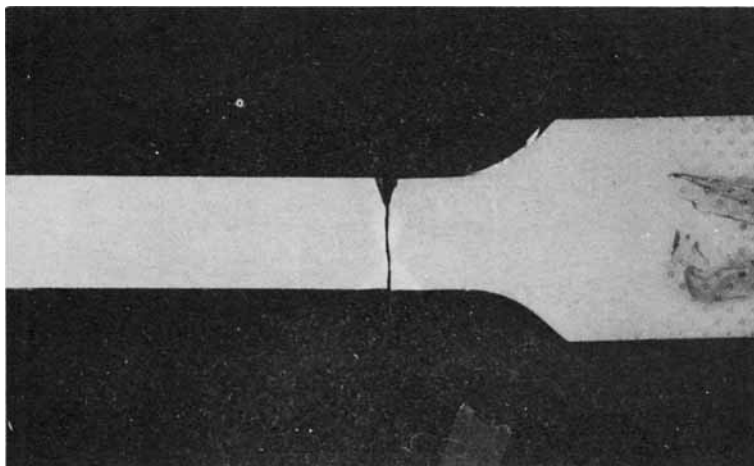


Fig. 17. Photograph of a fractured Instron specimen where failure appeared to occur at a "butterfly" pattern.

epoxy systems. Figure 17 shows a fractured Instron specimen where failure appeared to occur at a butterfly pattern.

Specimens fractured with the tensile impact test, at the highest amine concentrations, besides whitening, began to show shear banding and butterfly patterns. A butterfly pattern growing in the shear plane $45\text{--}50^\circ$ to the fracture surface has been observed in a few fracture specimens. Control samples also exhibit shear banding when fractured at a slow strain rate, but no obvious shear banding is observed in the tensile impact specimens. Sample observations and a summation of stress-strain results for the mode of failure study are presented in Table I.

Proposed Toughening Mechanism

Several theories exist for explaining the nature of toughening in rubber-modified compositions. For a theory to be acceptable, it must be able to account for the effects of structure upon fracture resistance, including the effects of rubber-matrix adhesion, rubber particle size, and the relaxation behavior of the rubber and matrix. The toughening theories must also explain the following mechanical behavior phenomena, on the basis of brittle glassy polymers: yielding, high elongation at break, high energy to break in impact, and stress whitening.

Toughening mechanisms for the rubber-modified epoxies can be divided into two categories: (a) the secondary rubber phase is considered as the major energy absorber and (b) the ability of the matrix to absorb energy, as a result of massive crazing (or simple voiding and microcavitation) and shearing. Only a few of the more recent theories are presented and evaluated in the following discussion.

The theory of Kunz-Douglas et al.,⁵ which proposes that toughening in rubber-modified epoxies is a result of rubber particles bridging the gap and stretching as the crack propagates, is one of the few theories which proposes that the rubber is the crucial parameter in the toughening process. The theory, which introduces some interesting ideas and analytical relationships, unfortunately was derived from low strain rate cleavage tests and

TABLE I
Comparison of Failure Mode for Samples Fractured at Slow and Fast Strain Rates^a

% excess amine	Wt % rubber	Stress-strain curve		Sample observation	
		Instron	Ten. impact	Instron	Ten. impact
0	0	pl	br	br	br
0	5	pl	br	br	br
0	10	pl	pl	br	w
0	20	pl	pl	br	w
20	0	pl	br	br	br
20	10	pl	pl	br	w
40	10	—	ss	—	w
50	0	ss	ss	n, sb	d
50	10	—	ss	—	w
60	0	ss	ss	n, sb	d
60	10	ss	ss	n, sb	2
70	0	—	ss	sb	sb
70	5	ss	ss	n	w
70	10	—	ss	—	sb, w, t, bu
70	20	ss	ss	n, sb, bu	sb, w, bu
80	0	ss	ss	sb, d	yd
80	10	ss	ss	n, sb, bu	w, bu, t
100	0	ss	br	sb, t	yd
100	10	ss	ss	sb, bu, t	sb, w, t, bu

^a br = brittle, pl = plastic deformation, ss = strain softening, t = tearing, sb = shear band, w = whitening, n = neck, bu = butterfly, and yd = yield deformation.

extrapolated to higher strain rates. This theory also does not take into account, nor explain, the stress whitening phenomenon at the high strain rates and the shearing at the lower strain rates, which has been reported in this investigation. The results presented here suggest no evidence to support that the stretching rubber accounts for more than a very small amount of energy absorption in a tightly crosslinked system, especially where plastic deformation is limited.

One of the original rubber-toughening theories, by Sultan and McGarry,⁶ dealt with the rubber as the major source of energy absorption. In this theory, the size of the rubber particle is the crucial factor. Large (1–5 μm) precipitated rubber particles tended to favor tensile crazing, while smaller particles (<0.5 μm) induced shearing. Epoxies modified with a combination of both types of particles⁶⁻⁸ were reported to exhibit maximum toughness, revealing both a crazing and shearing energy absorbing mechanism. This theory, though accounting for the observed results, does not take into account changes in the viscoelasticity of the matrix. The theory is derived from a ductile epoxy system (T_g around 90°C) with very low strain rate testing. It does not predict how the rubber particle size will affect toughening in a stiffer, high T_g epoxy network.

Pearson and Yee⁹ have proposed that toughness enhancement for rubber-modified epoxies results from the blunting of the sharp crack by cavitation of the rubber particles, which dissipates bulk strain energy and shear band formation promoted by the particles or cavities, which, in turn, dissipates more strain energy. Fracture toughness was reported to be more a function of rubber content than rubber particle size. Although Pearson and Yee are

correct in assuming that both cavitation and shear banding are major energy absorbing mechanisms, except for the most ductile systems, these two toughening processes have not been observed to occur simultaneously in a specimen fractured at a constant rate. Shear banding, which requires a greater amount of plastic deformation, tends to occur more often at the slow strain rates. At high strain rates, cavitation occurs, and, except for the very ductile systems, the matrix chains do not have enough time (relaxation time too high) to respond to the load; thus shearing is not observed. The theory of Pearson and Yee, though being perfectly correct, can only be applicable for toughening in a highly flexible system. The theory does, however, point out that intrinsic matrix ductility is the main source of toughness. This suggests that the more ductile the matrix is, the more it can be toughened.

Manziona, Gillham, and McPherson¹⁰ have proposed a stress response model which takes into account rubber-epoxy compatibility, the degree of interfacial bonding, and matrix viscoelasticity, using an extension of the Ludwik-Davidenkow-Orowan hypothesis.¹¹ A determination of the mode of material failure is made for a given rubber-modified epoxy at a given rate of deformation, using simple stress vs. rate of test and stress vs. temperature curves for debonding and yield stress.

From theory, in a rubber-modified composition, there are three competing mechanisms of failure: yielding (as evidenced by shear banding and necking), stress whitening (debonding, microcavitation, and crazing), and brittle failure. Whichever mechanism occurs at the lowest stress level will be the observed mechanism. The initiation energy for all three mechanisms will vary with temperature of test and rate of strain. The yield stress involves a viscous dissipation mechanism and is assumed to show the greater temperature and rate sensitivity. For the controls (also one-phase systems), there exist only two response mechanisms, brittle fracture and yielding. Again, whichever mechanism occurs at the lowest stress level will be the observed mechanism. In all cases, the mode of failure is controlled by the matrix viscoelasticity.

To accommodate the results reported in this article, the toughening theory of Manziona et al. must be modified somewhat. In the modified theory, the role of the dissolved rubber phase will not be dealt with as a major contributor toward toughening. Enhanced matrix ductility will come from reducing the network crosslink density, by using greater concentrations of amine curing agent.

For epoxy compositions, both rubber-modified and control, at amine concentrations close to stoichiometric equivalence, the epoxy matrix is brittle. The brittle fracture stress is lower than the yield stress for low and high strain rates. The failure mechanism for both the controls and rubber-modified compositions will be brittle fracture (Fig. 18). At higher amine concentration, the epoxy network is more flexible, and the yield stress is lowered. For the control specimen, failure will occur by either a brittle or yielding mechanism, depending on the amine concentration and the rate of strain. The transition point seems to occur for the MDA control formulations at 50% excess amine, at both high and low strain rates. At this amine concentration, evidence of a yield point is present in both Instron and tensile impact test specimens.

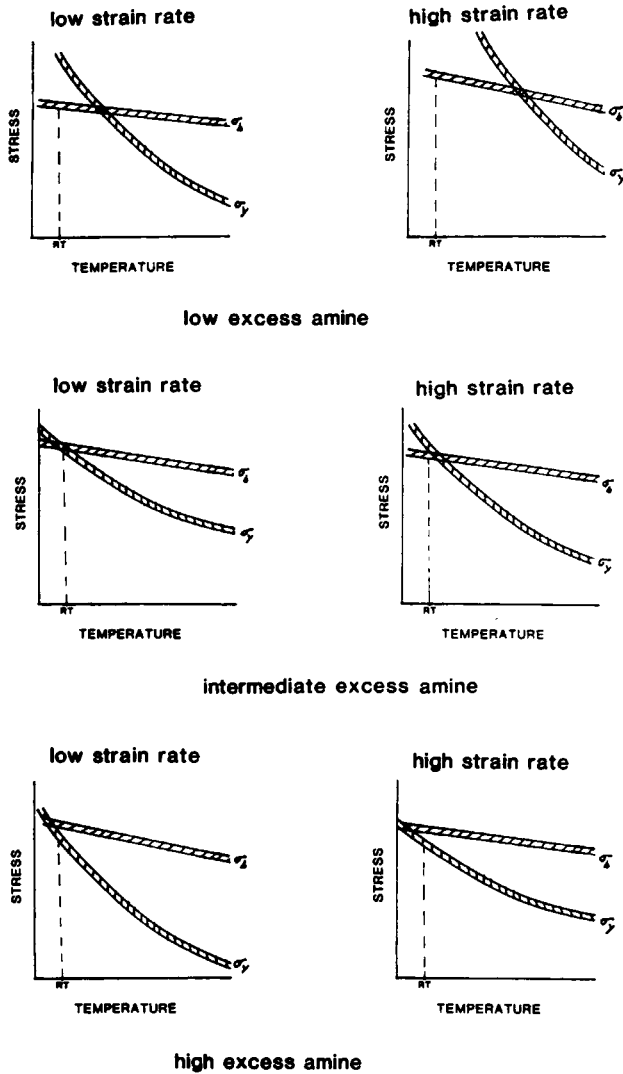


Fig. 18. Mechanism of failure for MDA-cured epoxy controls.

The rubber-modified compositions, at intermediate amine concentrations, exhibit debonding (whitening) when fractured at high strain rates, and matrix yielding at the slower rates. The stress curves for these two situations are shown in Figure 19. At slow test rates, the yield stress is lowered below the debonding stress. Evidence for this comes from the Instron fractured specimens. At the slow strain rate, samples yielded by necking or shearing near the fracture surface, with little evidence of whitening. Conversely, the entire gauge length of the tensile impact specimen whitened during failure and yielding was unobserved, or masked by the debonding phenomenon. At high amine concentration, the yield stress is further lowered, and, at some point, yielding will occur even at the high strain rates.

For the MDA system, unlike the one used by Manzione et al., the presence of the dissolved rubber does not appear to improve the energy to break at

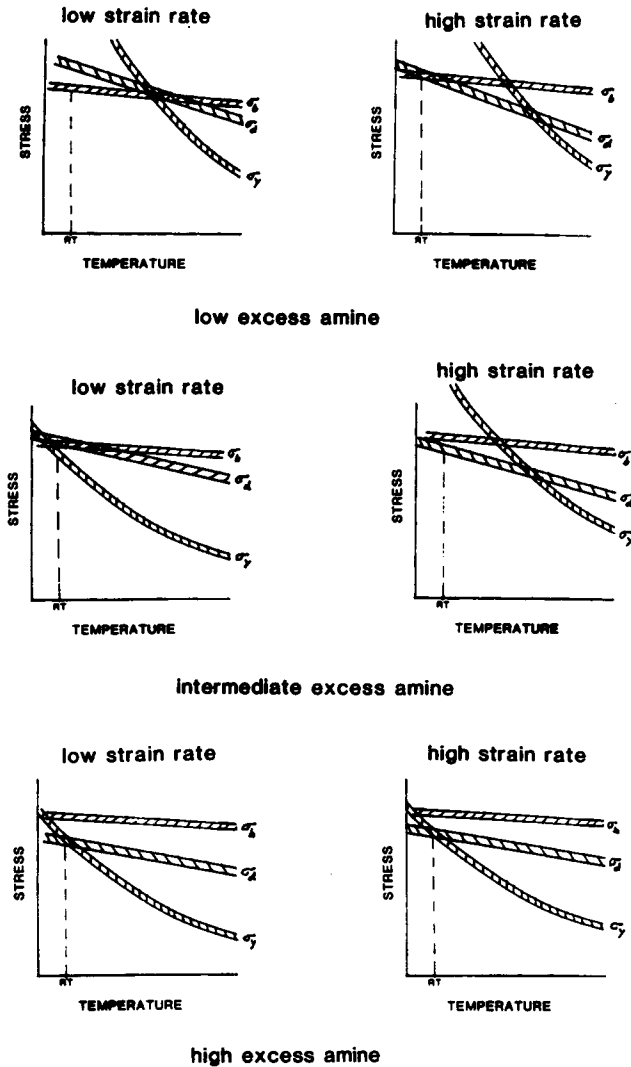


Fig. 19. Mechanism of failure for rubber-modified MDA-cured epoxy.

the lower strain rates. The elongations to break in the rubber-modified compositions are not much greater than the controls. Manzione et al. reported large differences in elongation between the controls and rubber-modified samples. This discrepancy might be explained by the fact that samples in Manzione et al.'s study were fractured at 1/10 of the rate of Instron speeds reported here. This difference in testing rate (even at such low rates) is quite large and could easily account for the lack of apparent gross yielding in the MDA system.

The rubber-modified compositions fractured at the high strain rates also lacked large improvements in elongation to break. The occurrence of stress whitening did not seem to have a major influence in improving toughening. One explanation given by Manzione et al. is that, even though stress whitening is an energy dissipating mechanism, the elongation in a sample is

limited, since the cavities, which compose the whitened region, quickly coalesce to produce catastrophic failure. Elongation to break becomes limited by the speed with which the cavities coalesce.

Up to this point, little has been said about the importance of the rubber in the toughening mechanism. From the experimental results, it appears that, for the proposed mechanism to be valid, the properties of the rubber and its initial compatibility and interfacial bonding with the epoxy are important. If the properties of the rubber are poor or the rubber-epoxy interfacial bonding is weak, crack propagation will occur prematurely (a crack propagates through a weak rubber or around a poorly bonded rubber), and the samples will break well below the yield or debonding stress. Having strong, initially compatible rubber, with good interfacial bonding does not always insure superior toughening. At amine concentrations approaching stoichiometric equivalence, brittle failure dominates, and the rubber acts more as a stress concentrator than as a toughening agent.

In summary, the toughening mechanism proposed here suggests that for a rubber-modified, two-phase system, the viscoelasticity of the matrix is the governing factor in the toughening process. The matrix viscoelasticity, in turn, is governed by the crosslink density of the matrix, strain rate and temperature of test, and the amount of rubber dissolved in the matrix. For the proposed mechanism to be valid, however, the properties of the rubber, its initial compatibility and interfacial bonding with the matrix, must be accounted for.

CONCLUSION

To better understand the role of the matrix viscoelasticity, control and rubber-modified compositions were formulated at different amine concentrations. The network crosslink density was lowered using larger amine concentrations. This allowed greater network mobility, with a shorter average relaxation time in response to an imposed stress. Impact strengths for the control and rubber-modified compositions increased with increasing amine concentration up to 70% excess amine (for the MDA system). These compositions had a greater capacity for absorbing energy. At amine concentrations greater than 70% excess, the matrix strength decreased. In all cases, the addition of the rubber did not produce a substantial impact strength improvement. Addition of a second rubber type, with poorer interfacial bonding capabilities, resulted in lower impact strengths at the higher amine concentrations. This confirmed that the choice of rubber is important in the toughening process, but, even with a "good" rubber, toughening improvements in a highly crosslinked, high- T_g , epoxy system are difficult to obtain.

A modified stress response model, first proposed by Mazione, Gillham, and McPherson, was used to fit the experimental results and to explain the toughening phenomenon. The model incorporates the rubber-epoxy compatibility, including the degree of interfacial bonding and matrix viscoelasticity.

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