Mechanical Properties of the Rubber-Toughened Polymer Blends of Polycarbonate (PC) and Poly(Ethylene Terephthalate) (PET)

ZHI-LI LIAO and FENG-CHIH CHANG*

Institute of Applied Chemistry, National Chiao-Tung University, Hsinchu, Taiwan, Republic of China

SYNOPSIS

The intrinsically impact-brittle PC/PET blends can be effectively toughened, in terms of lower ductile brittle transition temperature (DBTT) and reduced notch sensitivity, by incorporating butylacrylate core-shell rubber. The rubber particles are distributed exclusively in the PC phase. Varying the PC melt flow rate (MFR) is more important than varying the PET I.V. to vary the low temperature toughness of the blends. PC with MFR = 3 is essential to produce the toughest PC/PET/rubber blend. The presence of rubber slightly relieves the strain rate sensitivity on yield stress increase. Lower MFR PC in the blend results in smaller activation volume and, therefore, higher strain rate sensitivity, because a greater number of chain segments are involved in the cooperative movement during yielding. Two separate modes, localized and mass shear yielding, work simultaneously in the rubber toughening mechanism. The plane-strain localized shear yielding dominates the toughening mechanism at lower temperatures and brittle failure, while the plane-stress mass shear yielding dominates at higher temperatures and ductile failure. The critical precrack plastic zone volume has been used to interpret the observed phenomenon. © 1994 John Wiley & Sons, Inc.

INTRODUCTION

Polycarbonate (PC) is an amorphous tough thermoplastic with balanced thermal and mechanical properties, however, PC is considered to be relatively poor in terms of solvent resistance. Poly(ethylene terephthalate) (PET) is also a mechanically tough (except for being extremely notch brittle), crystallizable thermoplastic, with good solvent resistance. Both PC and PET are widely used commercial products and polymer blends of PC and PET have attracted great attention since the first report appeared in a U.S. patent.[1] Blending PET with PC improves the PC solvent resistance properties and reduces the final product's cost relative to PC.

Miscibility and the ester-interchange reaction between PC and PET have been the subjects of extensive investigations in the literature.[2-14] Studies, emphasizing the mechanical properties of PC/PET blends, except for a few cases,[15-17] are mainly concentrated in the patent literature.[18-26] Polymer blends of PC/PET are brittle, relative to PC, and an additional impact modifier (rubber) is usually required to improve the toughness of the blends to be practically useful. One patent stated that higher I.V. (intrinsic viscosity) of PET alone, without the presence of impact modifier, was able to provide a tough PC/PET blend.[27] The first reported patent on the impact-modified PC/PET blend was issued in 1975, using methyl methacrylate-butadiene-styrene (MBS) core-shell elastomer as an impact modifier.[18] Since 1975, a number of types of rubbers have been used to toughen PC/PET blends.[19-26]

The added rubber particles are usually distributed in the PC phase in PC/PET (or PC/PBT) blends, because most rubber outer structures are more compatible with PC than with PET or PBT.[28-30] We have been able to control the final rubber destination in PC/PET blends (in PC, PET, or in both phases) by selecting rubbers containing reactive functional

* To whom correspondence should be addressed.

Journal of Applied Polymer Science, Vol. 52, 1115-1127 (1994)
 CCC 0021-8995/94/081115-13

groups.[31] We also discovered that rubber, distributed in the PC phase, is more effective than that distributed in the PET phase for toughening of the blends.[31]

In our previous article,[17] we reported on rubber-toughened PC/PET blends using a higher PC content (PC/PET = 70/20 and 45/45) in the blends. In this article, we concentrate on the blends with PET being the major component (PC/PET = 35/55), which is close to most commercially available products.

EXPERIMENTAL

Materials

Natural grade PCs with various melt flow rates were obtained from Dow Chemical Company. PET samples, I.V. = 1.0, 0.92, and 0.84, were kindly donated by Far Eastern Textile Ltd. of Taiwan. Coreshell rubber, EXL 3330, with butylacrylate core and methyl methacrylate shell, was obtained from Rohm & Haas.

Melt Blending and Injection Molding

Melt blending was performed using a 20 mm welding engineering twin-screw counter-rotating extruder with L/D = 48. Both PC and PET pellets were dried in an oven at 120°C for 12 h and the rubber was dried at 70°C for 12 h prior to extrusion. The extrusion temperature was maintained at 260–265°C. The screw rotating rate was set at 230 rpm unless it was a testing variable. The blended pellets were dried again at 100°C for 10 h and were molded into test specimens on an Arburg 3 oz injection molding machine. The injection pressure was maintained at 75 (machine full scale at 99), unless it was treated as a testing variable.

Melt Flow Rate Measurements

The melt flow rate of the blended pellets was measured at 265°C and at 2.16 Kg loading.

Izod Impact and Tensile Tests

Notching of the $\frac{1}{8}$-in. specimens was carried out using a single tooth cutter at ambient conditions with a radius of 2.5, 10, and 20 mil. Izod impact tests of various temperatures were performed in a TMI impactor, equipped with a temperature-controlled chamber, which could be operated from −100 to 150°C. Standard tensile (ASTM D 638) and flexural tests (ASTM D 790) were carried out by an Instron universal testing machine, model 4201. In order to obtain the rate-dependent yield stress and modulus, tensile tests were also conducted by changing the crosshead speed from 1 to 500 mm/mim.

Scanning Electronic Microscopy (SEM) and Transmission Electron Microscopy (TEM)

Scanning electronic microscopy of the fractured surfaces was performed on a Hitachi S-570 SEM after the specimens were sputter-coated with gold. Small pieces, cut from the section perpendicular to flow direction, were microtomed, were stained with OsO_4 solution, and were examined by TEM on a JEOL 2000 TEM.

RESULTS AND DISCUSSION

Melt Flow Rate

The summarized MFR data are shown in Table I. The blend, containing higher MFR of PC, also has higher MFR, even when PC is the minor component in the blend. The presence of rubber causes a decrease in MFR, as would be expected. The observed MFR remains nearly constant for the blends containing PET with different I.V.

Izod Impact: Effect of Extruder Rotating Rate (RPM)

Figure 1 show the Izod impact strength vs. temperature by varying extruder rpm. The observed ductile brittle transition temperatures (DBTT) are essentially independent of the extruder rotating rpm with little variation of the impact strength. Ductile brittle transition is a change in fracture behavior from ductile to brittle or brittle to ductile, in response to the change of a variable during testing. Many reasons have been suggested to explain the phenomenon of ductile brittle transition in polymers, and it seems clear that there is no overriding mechanism that is responsible for this behavior. Pitman et al.[33] have accounted for the phenomenon observed in polycarbonate, in terms of competition between shear yielding and crazing, with shear yielding promoting ductile behavior and crazing causing brittle fracture. Greater mixing rate, achieved by higher extruder rpm, appears to be offset by the shorter mixing time and, therefore, the effect of extruder rpm on the resulting product toughness, within the range of our study (150–300 rpm), is insignificant.

Table I Summarized Data on MFR, Tensile, Flexural, and Activation Volume

Composition	MFR (g/10 min)	Tensile Yld. Str. (MPa)	Tensile Elong. (%)	Flexural Modulus (MPa)	Act. Vol. ν^* (nm^3)
PC3/PET/EXL = 35/55/0	20.5	65.1	79	2500	2.21
PC3/PET/EXL = 35/55/2.5	19.9	60.5	46	2410	2.21
PC3/PET/EXL = 35/55/5	21.3	57.8	35	2190	2.43
PC3/PET/EXL = 35/55/10	13.9	51.5	51	2060	2.43
PC3/PET/EXL = 35/55/15	11.6	47.4	53	1940	3.00
PC3/PET/EXL = 35/55/20	9.9	43.8	41	1780	3.62
PC10/PET/EXL = 35/55/0	23.3	—	—	2560	—
PC10/PET/EXL = 35/55/10	17.7	—	—	2230	—
PC15/PET/EXL = 35/55/0	26.0	64.1		2530	2.90
PC15/PET/EXL = 35/55/10	21.4	49.1	—	2080	2.48
PC20/PET/EXL = 35/55/0	26.9	62.5	—	2580	3.13
PC20/PET/EXL = 35/55/10	22.2	49.8	—	2040	3.51
PC80/PET/EXL = 35/55/10	37.6	—	—	—	—
PC3/PET*/EXL = 35/55/10	14.4	—	—	1990	—
PC3/PET**/EXL = 35/55/10	14.1	—	—	2000	—

All PET are I.V. = 1.00 except PET* = 0.92 and PET** = 0.84.

Izod Impact: Effect of Injection Pressure

Figure 2 shows a significant effect of the injection pressure on the product toughness in terms of DBTT. Higher injection pressure means higher injection flow rate (higher shear) during filling the cavity of the molder. This will result in greater specimen orientation and will become tougher in the direction perpendicular to the flow of the specimen, as would be expected.

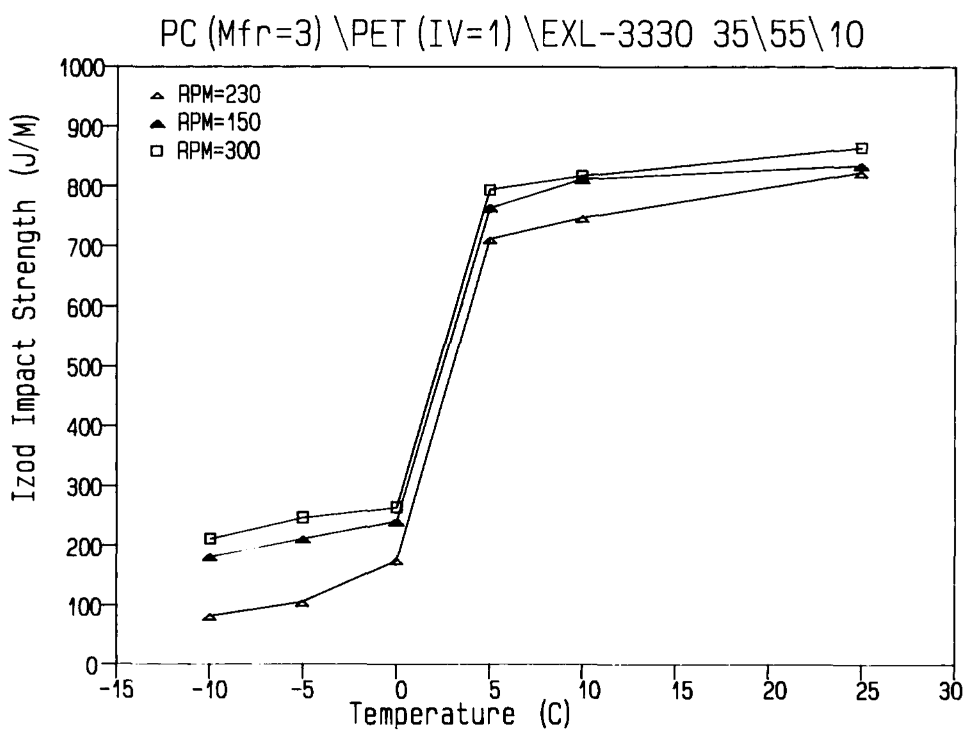

Figure 1 Effect of extruder rotating rate.

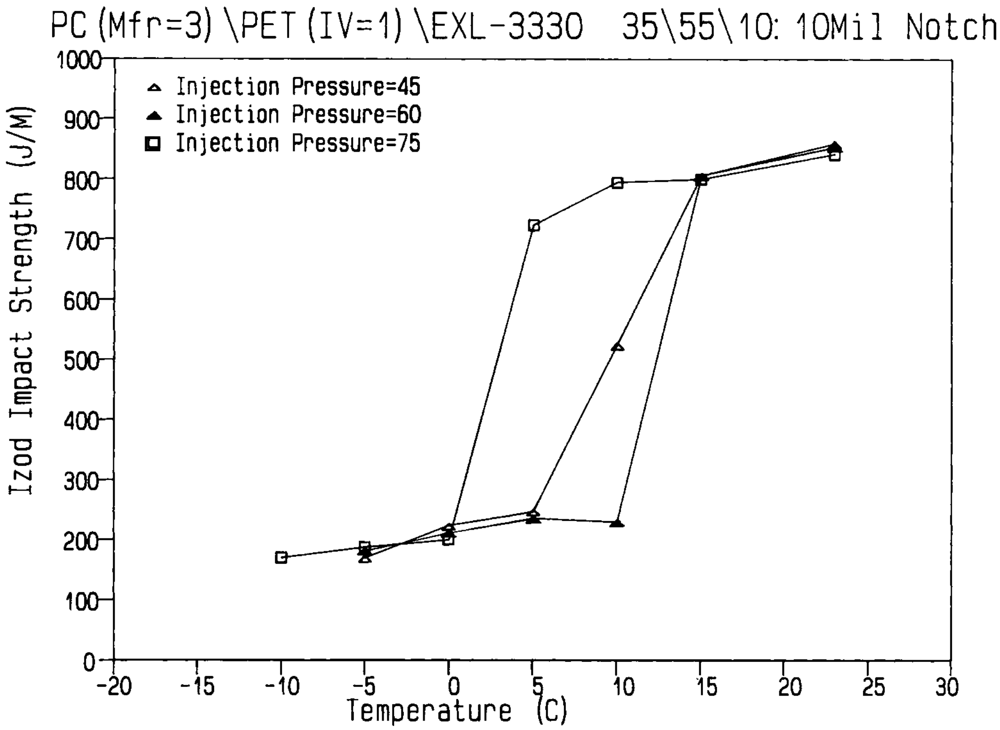

Figure 2 Effect of injection pressure.

Izod Impact: Effect of PC MFR and PET I.V.

Figure 3 shows the standard 10 mil Izod impact strengths of the PC/PET/rubber = 35/55/10 blends by varying temperature and PC MFR. The blend with PC3 has the lowest DBTT, at about 0°C, while the blend with PC80 has the highest DBTT, at about 50°C. The blends with intermediate MFR

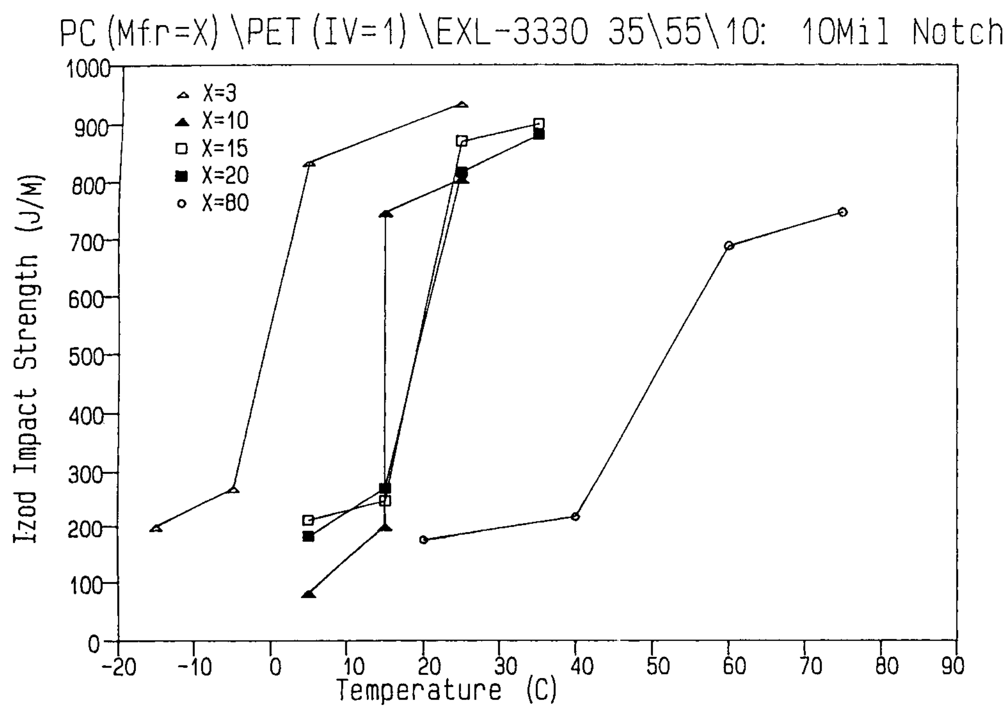

Figure 3 Effect of PC MFR in PC/PET/EXL blends.

have nearly identical DBTT around 15–20°C. PC MFR has been demonstrated to be important in dictating the toughness of the resultant blends, in terms of DBTT, even though PC is present as a minor component and a dispersed phase in these blends.

Figure 4 illustrates that the DBTT of the Izod impact strength is nearly independent of the I.V. of the PET component, within an I.V. range between 0.84 and 1.00. This is unexpected, because the PET is the dominant component and is the continuous phase in the blends, and should have a significant effect on the resulted product toughness. This observed result is contradictory to the previous claim that PET I.V. is the dominant factor in dictating the final toughness of the PC/PET blends.[27] PC is, therefore, a more important component than PET in deciding the final toughness of the resulting blend.

Izod Impact: Effect of Rubber Content

The presence of rubber tends to shift the DBTT lower, an indication of improvement in low temperature ductility. Figure 5 shows plots of impact strength vs. temperature with varying rubber content of the blends, PC/PET/EXL = 35/55/x. More rubber content in blend yields higher impact strength than does the lower content blend if the failure is in brittle mode, but has lower impact strength if the failure is in the ductile mode. Similar results were also observed in the PC/PET/rubber with higher PC content[17] and PC/rubber systems.[32] Blends with rubber content of 10 phr or greater all result in DBTT 0°C or below. DBTT can serve as a guide to determine the ductility of the blend, while the absolute impact strength is considered less essential if the fracture is ductile.[32] The unmodified blend, or the blends containing 5 phr or less of rubber, result in brittle failure at ambient.

Izod Impact: Effect of Notch Radius

Changing the specimen notch radius has a detrimental effect upon the impact behavior of essentially all the polymeric materials. The presence of a sharp notch not only creates a locally high triaxial stress concentration, but also increases the local rate of strain at the notch tip. The effect of changing notch radius, ρ_c, upon the impact strength can be rationalized in terms of stress concentration, η, at the root of the notch by the following equation,

$$\eta = 1 + 2(a/\rho_c)^{1/2}$$

where a is the notch depth and ρ_c is notch radius. Figure 6 shows the plots of impact strength vs. temperature of the blends, PC/PET/EXL = 35/55/x,

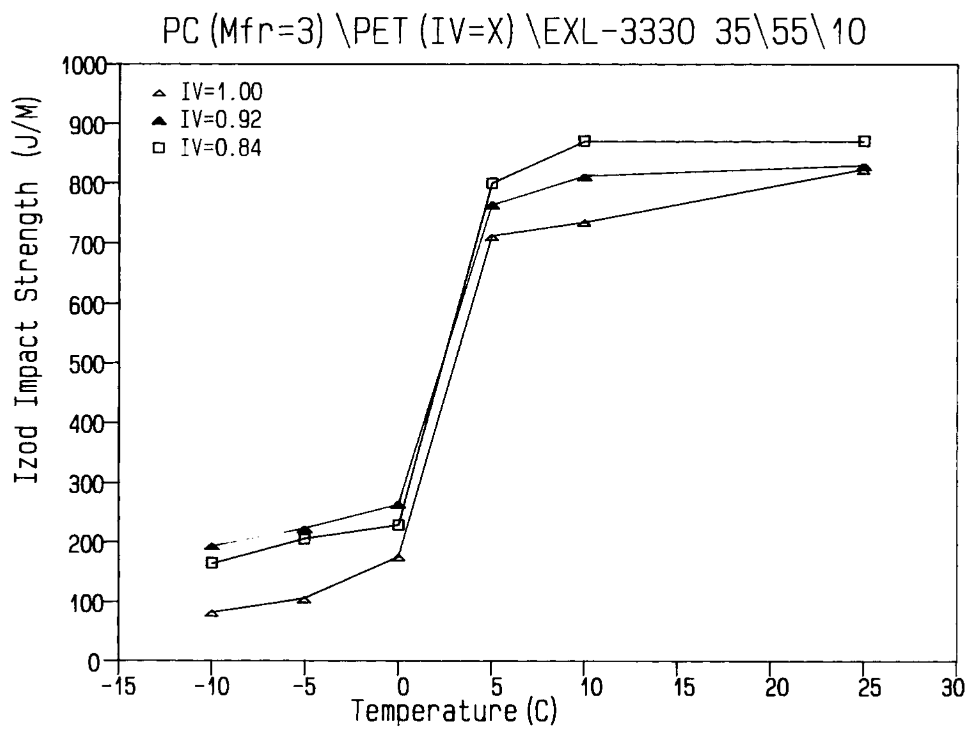

Figure 4 Effect of PET I.V. in PC/PET/EXL blends.

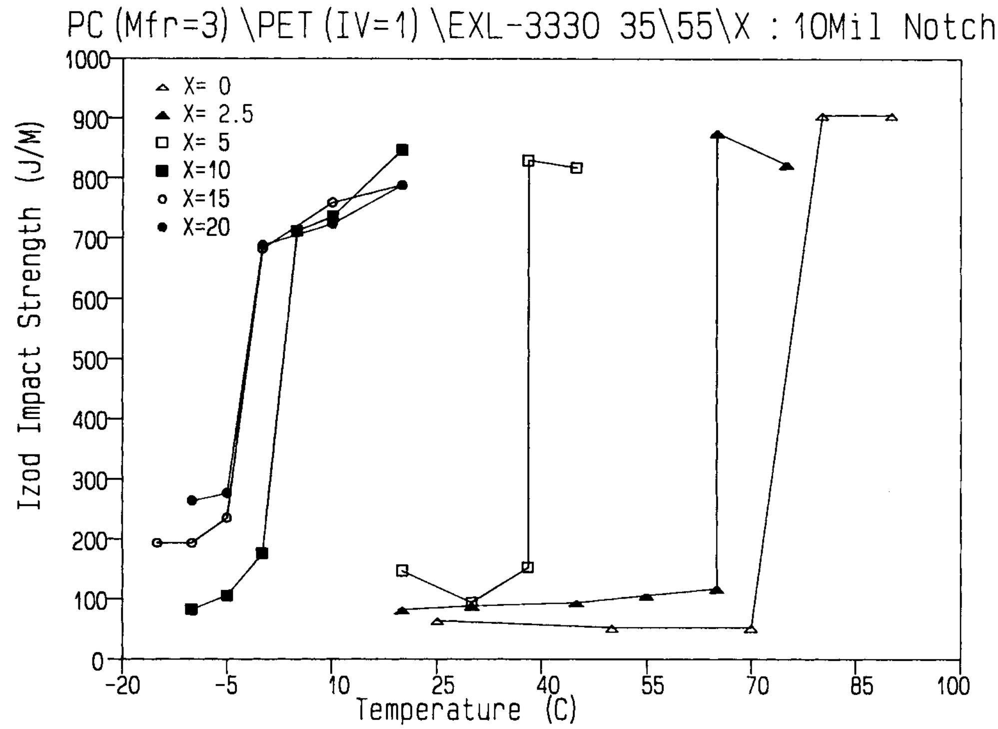

Figure 5 Effect of rubber content in PC/PET/EXL blends.

varying rubber content by using a smaller notch radius (2.5 mil). Relative to the results obtained from the standard 10 mil notch radius, the DBTT of the same composition is relatively higher, as would be expected. Figure 7 illustrates the effect of notch radius and rubber content on DBTT. Reduction of the DBTT, due to the presence of rubber, is especially pronounced for those blends with smaller notch ra-

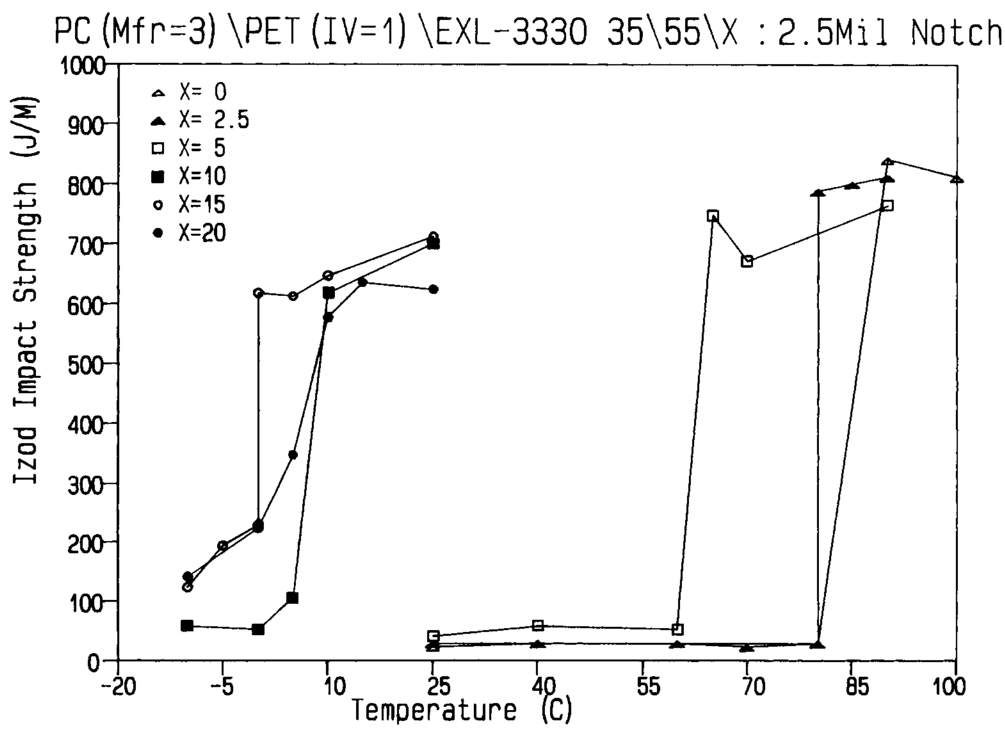

Figure 6 Effect of reduced notch radius in PC/PET/EXL blends.

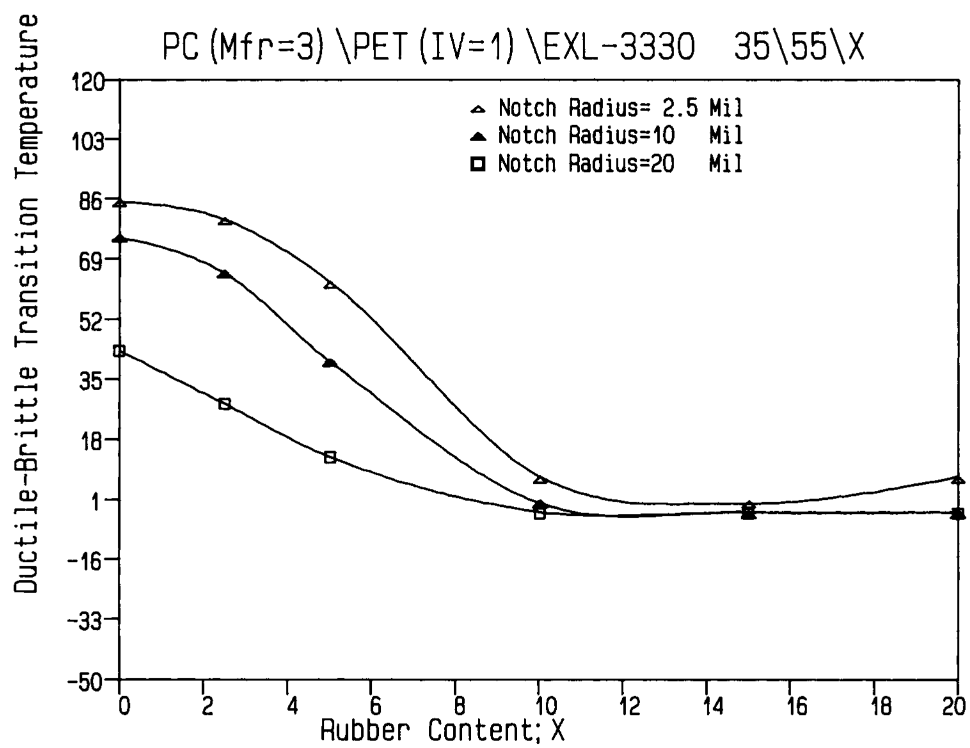

Figure 7 Effect of rubber content and notch radius on DBTT.

dii. The presence of rubber enables the reduction of notch sensitivity of the unmodified matrix (or blend) that has been demonstrated in this study.

Tensile and Flexural Properties

Table I summarizes the key tensile and flexural properties, including tensile yield stress, elongation to break, and flexural modulus at a crosshead speed of 50 mm/min. The yield stress and flexural modulus decrease progressively with the increase of rubber content, which is similar to most rubber modified plastics. Varying rubber content shows an insignificant effect on tensile elongation.

Tensile Yield Stress at Various Strain Rates

The typical Izod impact test has a hammer striking velocity of about 3 m/s. In the notched beam tests, the strain rate at the notch tip is considered significantly higher and has been estimated on the order of $5 \times 10^3\ s^{-1}$.[34] The yield behavior of glassy polymers is dependent upon temperature and strain rate. Bauwens-Crowet et al.[35] studied the dependence of polycarbonate yield stress on strain rate under different temperatures and they modeled this dependence using the Eyring theory of viscosity.[36]

$$\frac{|\sigma_y|}{T} = \frac{\Delta E^*}{v^* T} + \left(\frac{R}{v^*}\right) \ln\,(\dot{e}/A_E)$$

where R is the gas constant, v^* is the activation volume, and A_E is a constant. It is easily understood from the above equation that a higher strain rate will cause an increase in yield stress. This means that, under the same hammer impact rate conditions, the notch tip with the smaller notch radius will result in even higher local strain rate and, therefore, the corresponding yield stress. On the contrary, the breaking stress (craze stress) is normally rate and temperature independent.

As mentioned earlier, ductile brittle transition behavior can be interpreted as a competition between yielding and crazing during fracture and the rate-induced higher yield stress certainly will favor brittle failure. Such a time-dependent mechanical property in polymeric materials has been generally recognized. For many polymers, a plot of the yield stress against the logarithm of the strain rate is linear, so that the gradient $(d\sigma_y/d(\ln \dot{e}))_T$ is a constant. It is this gradient that determines the activation volume (v^*) from the above Eyring equation and the strain rate sensitivity is often discussed in terms of the activation volume. For PC, the activation volume was determined as 2.8 nm^3,[35] which is much greater than the volume of a statistical random link of about 0.5 nm^3.[37] It has been found that the values of v^* for most polymers were considerably greater than the volume of the statistical random link in the polymer chain in dilute solution, by a factor of about 2–10, depending upon the polymer.[37] This has been interpreted to mean that yielding involves the cooperative movement of a larger number of chain segments than would be required for a molecular conformation change in dilute solution.

There is not yet sufficient evidence for a true physical interpretation of v^*, and attempts to correlate v^* with specific features of the molecular structure have remained speculative. According to the above Eyring equation, material with higher v^* means less rate sensitivity of the yield stress increase. If the activation volumes of the blends, containing various quantities of rubber, can be determined, we can understand whether the presence of rubber is indeed reducing the rate-induced yield stress increase and raising the resultant toughness. Figure 8 shows the plots of the logarithm of strain rate vs. yield stress at ambient conditions for the blends PC/PET/EXL = 35/55/x. The activation volumes, calculated from the slopes of the straight lines, are in the same range as for the polycarbonate previously reported[35] and the results are summarized in Figure 9 and Table I. Figure 9 shows the increasing trend of the activation volume with the increase of rubber content, although the variation is considered insignificant and may be within experimental error. This means that the presence of rubber in PC/PET blends may slightly reduce the rate sensitivity of the yield stress increase. On the contrary, the more ductile blend, with higher PC molecular weight (lower MFR), results in a smaller activation volume (Table I). It would appear reasonable that the longer chain length of a higher MW PC would require an even greater number of chain segments to be involved in such cooperative movement during yielding.

SEM and TEM Morphologies

Morphology of a fracture surface, an indication of surface energy dissipations during fracture, is able to differentiate the relative ductility of polymeric

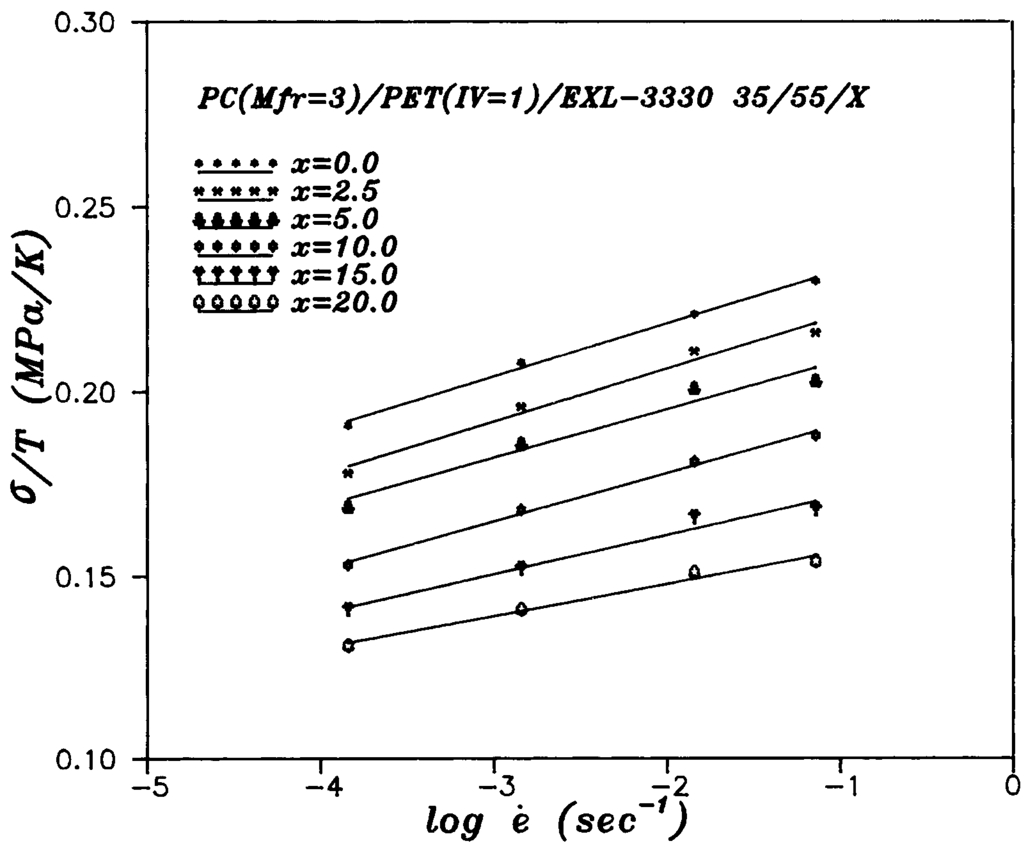

Figure 8 Plots of p/T vs. log e varying rubber content on PC/PET/EXL blends.

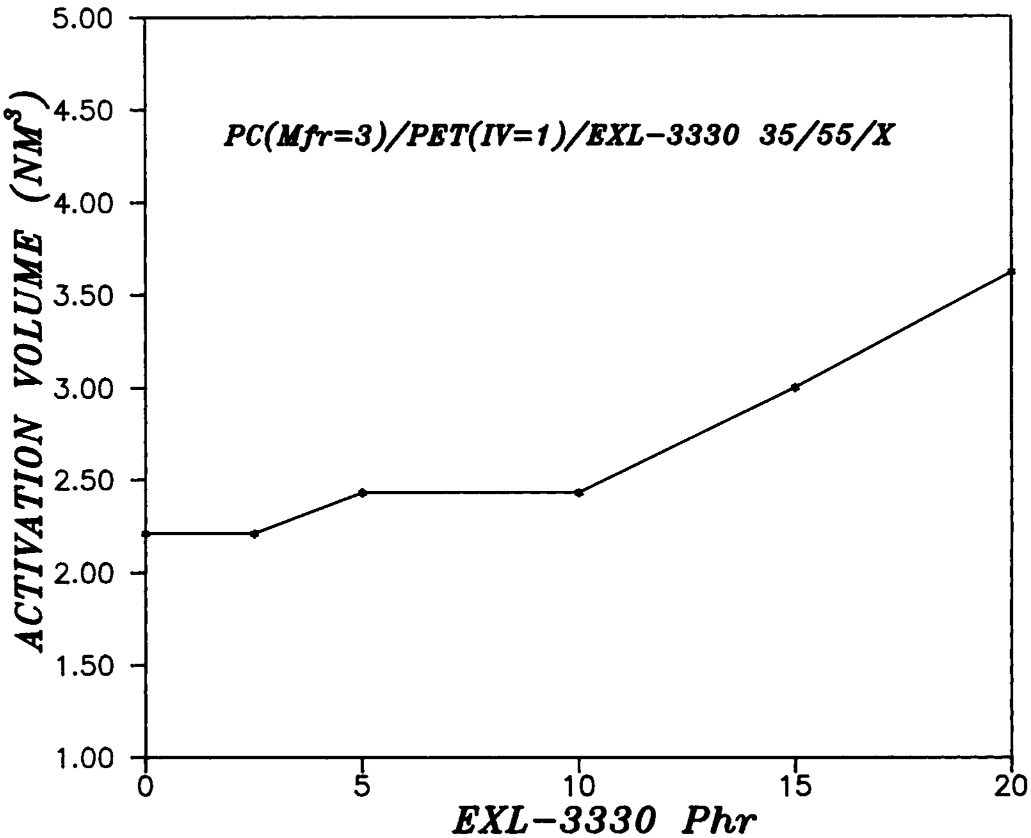

Figure 9 Effect of rubber content on activation volume.

materials in most cases. However, when the fracture is of a mass shear yield ductile mode, energy dissipation underneath the fracture surface dominates the total energy consumption and the localized energy dissipations from the surface become less important.[38] Figures 10(A–D) illustrate the fracture surfaces of the unmodified blend PC/PET/EXL = 35/55/0. The fracture surface of the ductile-mode specimen [Fig. 10(A)] clearly shows the lateral contraction, while the fracture surface of the brittle-mode [Fig. 10(C)] is relatively flat, without lateral contraction. At higher magnification, rougher surface morphology is also observed for the ductile-mode specimen than is observed for the brittle-mode specimen, as shown in Figures 10(B) and (D). Figures 11(A–D) show the typical fracture surfaces of the rubber modified blend PC/PET/EXL = 35/55/10, where the ductile-mode fracture surface of the blend also has the characteristic lateral contraction. The degree of lateral contraction is directly related to the observed impact strength if the failure is in a ductile mode.

Figures 12 and 13 show the TEM micrographs of the blends, PC/PET = 35/55 and PC/PET/EXL = 35/55/10, respectively. The relatively more ductile PC phase exists as a dispersed and elongated lamellar structure, while the PET is the continuous matrix. The butylacrylate rubber particles cannot be stained by OsO_4 and are shown as white particles distributed in the darker PC phase (Fig. 13). The shell structure of the rubber, PMMA, is relatively more compatible with PC than with PET, therefore the rubber particles are preferably distributed in the PC phase. Details on rubber distribution in PC/PET/rubber blends have been reported previously.[31]

Rubber Toughening Mechanism

Both PC and PET are considered to be pseudoductile polymeric materials, which characteristically have high impact strength on unnotched specimens, but low impact strength on notched specimens. The main emphasis in toughening these pseudoductile polymers is to promote the failure mechanism from the localized shear yield brittle mode to the mass shear yield ductile mode.[32] The PC component in the blends is a more important contributor than the PET component to resist notch fracture. The PC

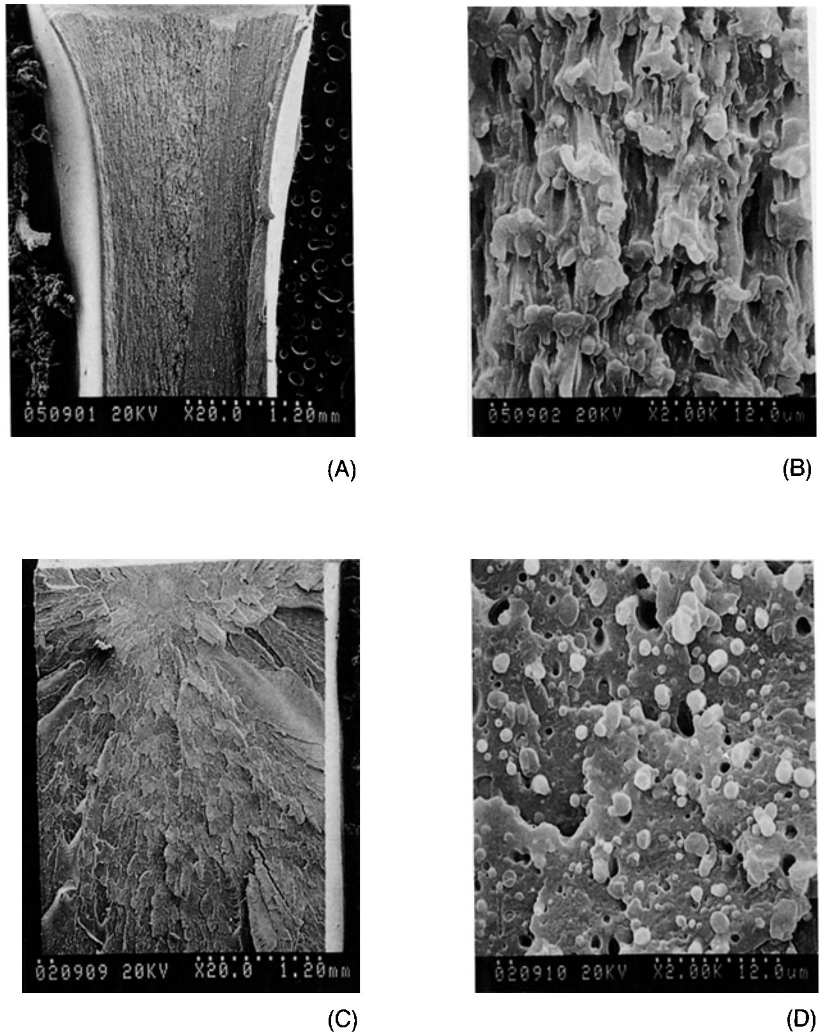

Figure 10 SEM photographs of PC/PET = 35/55 blend. (A) Ductile fracture at 80°C, ×20, (B) ductile fracture at 80°C, ×2000, (C) brittle fracture at 25°C, × 20, and (D) brittle fracture at 25°C, × 2000.

with lower MFR is relatively tougher in terms of lower DBTT than PC with higher MFR.[32,39] Similar results were also obtained in PC/rubber and PC/ABS blends.[32,39]

Rubber toughening of the PC matrix is considerably more effective than rubber toughening of the PET matrix in terms of lowering their DBTT.[17,32,38] Usually, PET requires 20% or more rubber to turn the notch brittle PET into a ductile product under ambient conditions. Our previous report indicated that an equal amount of rubber, distributed in the PC phase, is indeed more efficient in toughening PC/PET blends than in the PET phase.[31] This butylacrylate rubber with PMMA shell structure resides in the PC phase in the PC/PET/rubber blends in this study.

The presence of rubber above its T_g reduces yield stress of the blend and enhances both mass shear and localized shear yield energy dissipations. Our previously proposed critical precrack plastic zone model offers a simple explanation of the rubber toughening mechanism.[38,40–42] The presence of rubber in a matrix (or blend) reduces yield stress, which can effectively blunt the crack tip and resist crack initiation. This allows the precrack plastic zone surrounding the crack tip to grow above a critical value and a crack, developed later, will proceed within the plastic zone and will result in ductile tearing. The elongated PC lamellae (with or without rubber) are the main toughness contributors to resist crack initiation and growth. If the dispersed PC phase exists as spherical particles in the blend (such as a com-

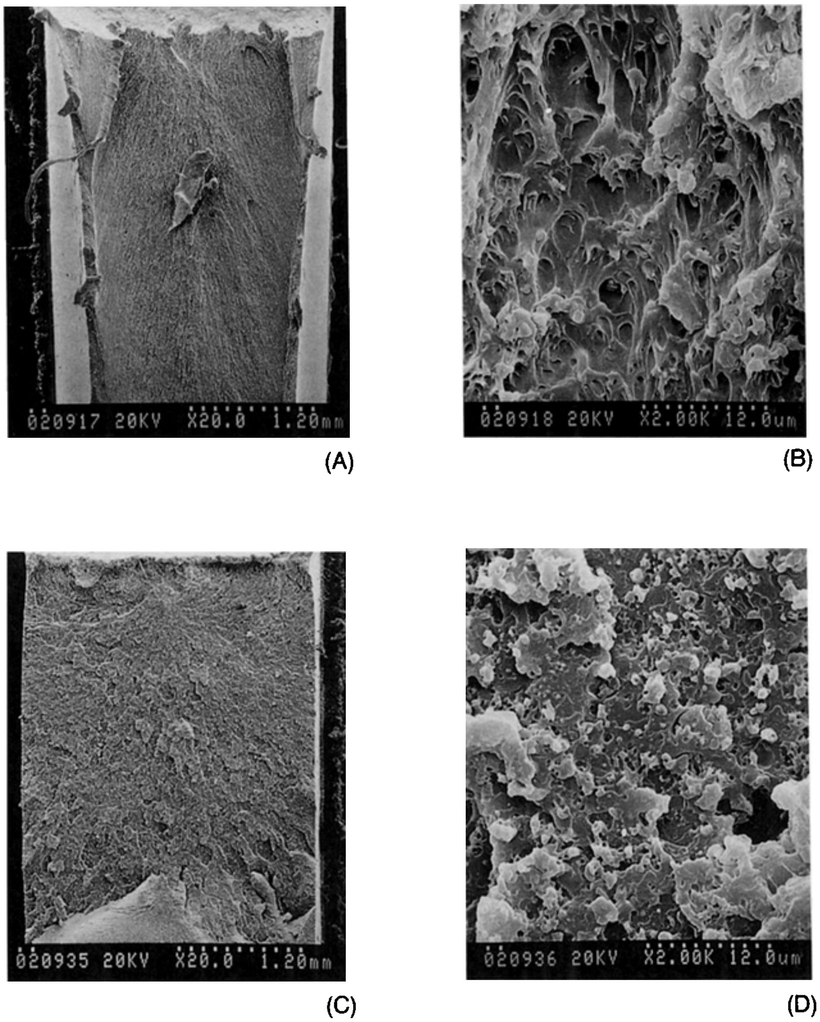

Figure 11 SEM photographs of PC/PET/EXL = 35/55/10 blend. (A) Ductile fracture at 25°C, × 20, (B) ductile fracture at 25°C, × 2000, (C) brittle fracture at −10°C, × 20, and (D) brittle fracture at −10°C, × 2000.

pression molded specimen), the crack growth can pass between PC particles. This would result in their being less effective in resisting crack initiation and growth and, therefore, the blend becomes less tough.

This study has demonstrated that the presence of rubber in the PC/PET blends does reduce the sensitivity of the strain rate-induced yield stress increase, but the difference is considered to be insignificant.

As mentioned above, the rubber modified blend has higher impact strength than does the unmodified counterpart, if the failure is in brittle mode, but has lower impact strength if the failure is in ductile mode. Therefore, the presence of rubber actually toughens the blend simultaneously in two separate modes, localized and mass shear yielding. The plane-

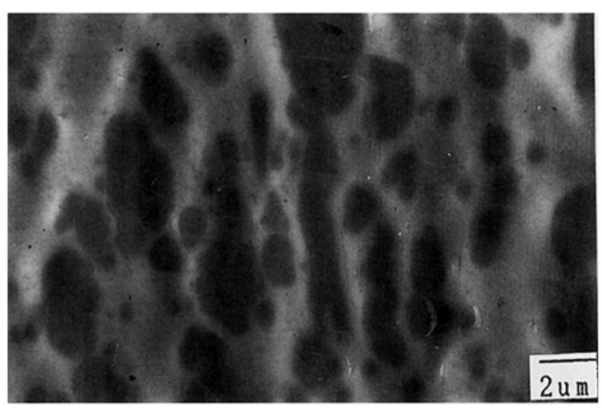

Figure 12 TEM micrograph of PC/PET = 35/55 blend.

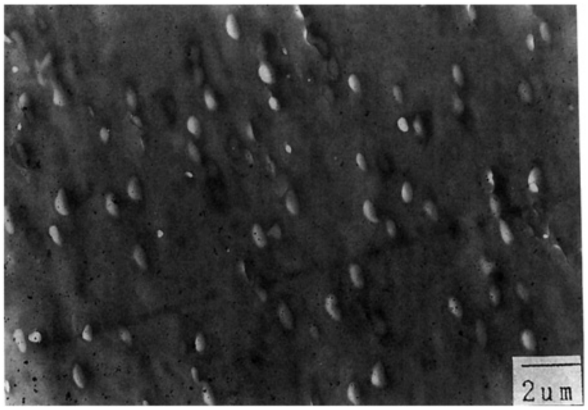

Figure 13 TEM micrograph of PC/PET/EXL = 35/55/10 blend.

strain localized shear yielding dominates the toughening mechanism at lower temperatures and brittle failure, while the plane-stress mass shear yielding dominates at higher temperatures and ductile failure. The criterion for shifting the fracture from brittle to ductile mode can be interpreted in terms of the precrack plastic zone volume we proposed previously, based on rubber toughening polycarbonates.[32,37,39,41]

CONCLUSIONS

The butylacrylate core-shell rubber, with PMMA shell structure, has been demonstrated to be effective in toughening polymer blends of PC and PET in terms of reducing DBTT. The butylacrylate rubber particles with PMMA shell are distributed exclusively in the PC phase. PC is a more important component than is PET in dictating the final toughness of the resultant blends. The presence of rubber may slightly relieve the strain rate induced yield stress increase, but the variation is essentially insignificant within experimental error. Lower MFR PC in the blend slightly increases the strain rate sensitivity yield stress increase, because it involves a greater number of chain segments in cooperative movement during yielding. Two separate modes, localized and mass shear yielding, work simultaneously in the rubber toughening mechanism. The plane-strain localized shear yielding dominates the toughening mechanism at lower temperatures and brittle fracture, while the plane-stress mass shear yielding dominates at higher temperatures and ductile failure. The critical precrack plastic zone volume can be used to interpret the observed DBTT phenomenon.

The authors express their thanks to the Integral Chemical Co., Ltd., of Taiwan and the National Council, Republic of China, for funding this research program. Thanks are also extended to the Far Eastern Textile Ltd. for providing free PET samples.

REFERENCES

1. U.S. Patent, 3,218,372, to Kunoshima of Japan (1965).
2. T. R. Nassar, D. R. Paul, and J. W. Barlow, *J. Appl. Polym. Sci.*, **23**, 85 (1979).
3. J. W. Barlow and D. R. Paul, *Polym. Eng. Sci.*, **21**, 985 (1981).
4. M. Droscher and F. G. Schmidt, *Polym. Bull.*, **4**, 261 (1981).
5. X. Y. Chen and A. W. Bireley, *Br. Polym. J.*, **17**, 347 (1985).
6. F. Pilati, E. Marianucci, and C. Berti, *J. Appl. Polym. Sci.*, **30**, 1267 (1895).
7. R. B. Hanrahan, S. R. Angeli, and J. Runt, *Polym. Bull.*, **14**, 399 (1985).
8. P. Godard, J. M. Dekoninck, V. Devlesaver, and J. Devaux, *J. Polym. Sci. Part A*, **24**, 3301, 3315 (1986).
9. R. B. Hanrahan, S. R. Angeli, and J. Runt, *Polym. Bull.*, **15**, 455 (1986).
10. Z. H. Huang and L. H. Wang, *Makromol. Chem. Rapid Commun.*, **7**, 255 (1986).
11. T. Suzuki, H. Tanaka, and T. Nishi, *Polymer*, **30**, 1287 (1989).
12. L. H. Wang, Z. Huang, T. Hong, and R. S. Porter, *J. Macromol. Sci. Phys.*, **29**, 155 (1990).
13. C. Berti, V. Bonora, M. Fiorina, and F. Pilati, *Makromol. Chem.*, **193**, 1665, 1679 (1992).
14. Q. Tan and D. Z. Ma, *J. Appl. Polym. Sci.*, **48**, 747 (1993).
15. S. R. Murff, J. W. Barlow, and D. R. Paul, *J. Appl. Polym. Sci.*, **29**, 3231 (1984).
16. J. Y. J. Chung, N. R. Lazear, and U. R. Grigo, *ANTEC 1985*, 944 (1985).
17. Z. L. Liao and F. C. Chang, *J. Polym. Res.*, to appear.
18. U.S. Patent 3,864,428, to Teijin (1975).
19. U.S. Patent 4,172,859, to Du Pont (1979).
20. U.S. Patent 4,267,096, to General Electric (1981).
21. U.S. Patent 4,522,979, to Mobay (1985).
22. U.S. Patent 4,554,315, to Mobay (1985).
23. J.P. 76. 39,749, to Mitsubishi (1976).
24. J.P. 83. 25,352, to Mitsubishi Chem. (1983).
25. J.P. 85. 127,352, to Mitsubishi Gas Chem. (1985).
26. J.P. 85. 127,353, to Mitsubishi Gas Chem. (1985).
27. J.P. 86. 43,656, to Kanebo (1986).
28. F. C. Chang, *Company Internal Report*, Dow Chemical Company (1985).

29. S. Y. Hobbs, M. E. J. Dekkers, and V. H. Watkins, *Polym. Bull.,* **17,** 341 (1987).
30. S. Y. Hobbs, M. E. J. Dekkers, and V. H. Watkins, *J. Mater. Sci.,* **23,** 1219 (1988).
31. K. P. Lin and F. C. Chang, *Polym. Networks & Blends,* to appear.
32. F. C. Chang, J. S. Wu, and L. H. Chu, *J. Appl. Polym. Sci.,* **44,** 491 (1992).
33. G. L. Pitman, I. M. Ward, and R. A. Duckett, *J. Mater. Sci.,* **13,** 2092 (1978).
34. F. J. Furno, R. S. Webb, and N. P. Cook, *J. Appl. Phys. Sci.,* **8,** 101 (1964).
35. C. Bauwnes-Crowet, J. C. Bauwens, and G. Homes, *J. Polym. Sci.,* **A2, 7,** 735 (1969).
36. H. Eyring, *J. Chem. Phys.,* **4,** 283 (1936).
37. R. N. Haward and G. Thackray, *Proc. Roy. Soc.,* **A302,** 453 (1968).
38. F. C. Chang and H. C. Hsu, *J. Appl. Polym. Sci.,* **47,** 2195 (1993).
39. J. S. Wu, S. C. Shen, and F. C. Chang, *J. Appl. Polym. Sci.,* **50,** 1379 (1993).
40. C. B. Lee and F. C. Chang, *Polym. Eng. Sci.,* **32,** 792 (1992).
41. F. C. Chang and H. C. Hsu, *J. Appl. Polym. Sci.,* **43,** 1025 (1991).
42. F. C. Chang and L. H. Chu, *J. Appl. Polym. Sci.,* **44,** 1615 (1992).

Received August 20, 1993
Accepted November 23, 1993