

Properties of PVC/CPE/EPDM Polyblends

YU-DER LEE and CHI-MING CHEN, *Department of Chemical Engineering, National Tsing Hua University, Hsinchu, Taiwan, Republic of China 30043*

Synopsis

A series of tricomponent blends of poly(vinyl chloride)/chlorinated polyethylene/ethylene propylene diene terpolymer (PVC/CPE/EPDM) were prepared and studied. CPE was used not only to improve the room temperature impact resistance of PVC but also as a polymer compatibilizer; while EPDM was used to enhance the impact resistance of PVC especially in low temperature range. Our data showed that the improvements of PVCs impact strength were significant either at room temperature or at low temperature (-12°C), however, a loss of the yield tensile strength was observed. On the basis of morphology, an impact-toughening mechanism of the tricomponent blend was proposed.

INTRODUCTION

Rigid poly(vinyl chloride) (PVC) has good mechanical properties, but two major disadvantages, namely, poor processability and low impact resistance.¹ A number of elastomer-based modifiers have been blended with PVC to improve its impact resistance.² However, due to its polar nature, PVC is not very compatible with most elastomers. Chlorinated polyethylene (CPE) is a low cost, semicompatible impact modifier with good weatherability; but due to its high glass transition temperature, it is unable to produce good properties modification at low temperature.³⁻⁶ On the other hand, ethylene/propylene/diene terpolymer (EPDM), with a low glass transition temperature, displays excellent low temperature impact resistance.^{7,8} Unfortunately, EPDM, being a nonpolar polymer, has poor interfacial adhesion with PVC; therefore they are essentially immiscible with each other. It is believed that by introducing a graft or block copolymer with segments similar to both plastic and rubbery phases will provide the necessary adhesion between the two phases.^{9,10} In this study, PVC, CPE, and EPDM are blended to form a tricomponent system. CPE, with ethylene segments similar to EPDM and chlorinated sequences similar to PVC,^{11,12} serves as a compatibilizer, as well as a high temperature impact modifier; while EPDM is chosen as the low temperature impact modifier.

EXPERIMENTAL

Materials

PVC chips were prepared by mixing 100 PHR PVC ($p_n = 800$), 3 PHR dioctylphthalate as plasticizer, and 4 PHR lead stearate as stabilizer in an

TABLE I
Characteristic Properties of CPE

Property \ CPE	CPE25	CPE36	CPE42
Chlorine content (%)	25	36	42
Residual crystallinity(%)	25	2	< 2
Density (g/c.c.)	1.1	1.16	1.22

extruder at 170°C and 40 rpm. The EPDM elastomer (Esprene EPDM 301, ethylene to propylene ratio = 63/35) was purchased from Sumitomo Chemical, Japan. The CPE impact modifiers used are products of Dow Chemical, U.S.A., their properties are listed in Table I.

Sample Preparation

Polyblends were mixed in a single-screw extruder at 170°C, 40 rpm, and pelletized. The chips were then injection molded to samples with standard dimensions according to ASTM D-256 and D-638, under an injection pressure of 90 kg/cm², at a barrel temperature of 175°C and a mold temperature of 70°C.

Measurement of Flex Temperature and Glass Transition Temperature

The flex temperature (T_f) was determined by a low temperature flexibility tester (torsional type) according to the testing procedures of ASTM D-1043. The glass transition temperature (T_g) was determined by a Rheovibron Viscoelastometer at 110 Hz.

Mechanical Testing

The yield tensile strength and break elongation at $25 \pm 2^\circ\text{C}$ were determined by an Instron Tester. The notched impact strength was measured at $25 \pm 2^\circ\text{C}$ and $-12 \pm 2^\circ\text{C}$ with an Izod Impact Tester. All specimens were allowed to stand at the testing temperatures for at least 24 h and then tested quickly to avoid temperature change during the test.

Electron Microscopy

Ultrathin sections for transmission electron microscopy (TEM) observation were prepared with a ultramicrotome at room temperature, then dipped in diazobicycloundecane (DBU) at 0°C for 24 h to introduce double bonds in CPE chain.¹² The DBU-treated specimens were then stained with 1% aqueous osmium tetroxide in room temperature for 2 h.

TABLE II
Flex Temperature (T_f) and Glass Transition Temperature (T_g) of CPE and EPDM

EPC T_f or T_g	CPE42	CPE25	CPE36	EPDM
T_f ($^{\circ}$ C)	-15	-19	-24	-56
T_g ($^{\circ}$ C)	—	—	15	-26

RESULTS AND DISCUSSION

Thermal Properties of CPE and EPDM

The flex temperatures (T_f) and glass transition temperatures (T_g) of CPE and EPDM measured are listed in Table II. The Flex temperature of EPDM is much lower than those of CPE. The glass transition temperature of EPDM is also much lower than that of CPE36. These thermal properties indicate that EPDM is more flexible than CPE at low temperature, and therefore can be a much better impact modifier under these conditions.

Properties of Bicomponent Polyblends (PVC/EPDM and PVC/CPE)

The yield tensile strength of both PVC/CPE and PVC/EPDM polyblends, illustrated in Figure 1, decreases as PVC composition increases. Moreover, higher chlorine content in the CPE causes less reduction in yield strength.

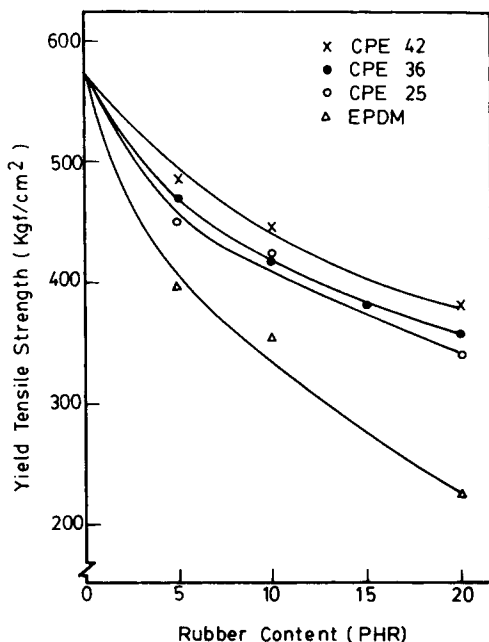


Fig. 1. Yield tensile strength of PVC/CPE and PVC/EPDM polyblend.

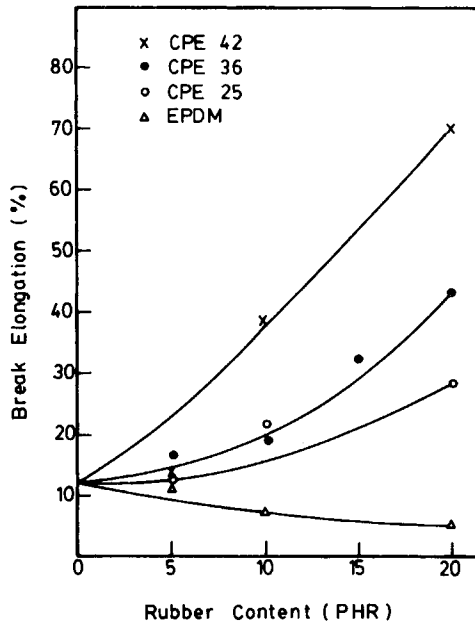


Fig. 2. Break elongations of PVC/CPE and PVC/EPDM polyblend.

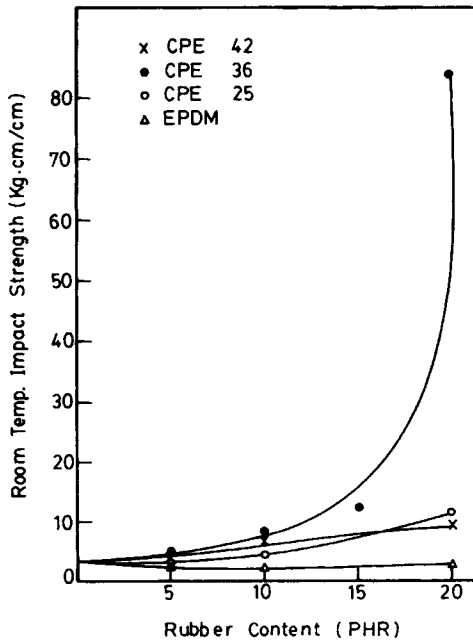


Fig. 3. Room temperature notched impact strengths of PVC/CPE and PVC/EPDM polyblend.

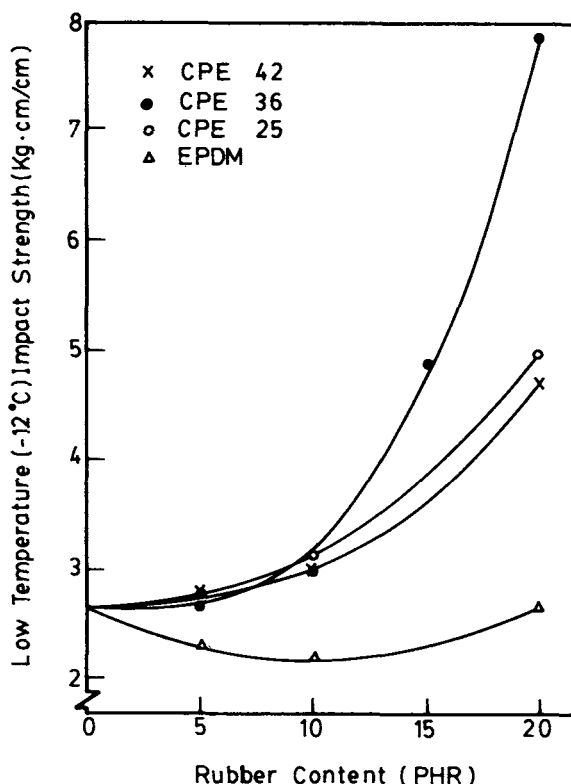


Fig. 4. Low temperature (-12°C) notched impact strengths of PVC/CPE and PVC/EPDM polyblend.

Figure 2 shows that break elongation of PVC is decreased by the addition of EPDM, but increases with CPE. We concluded that the interfacial adhesion between EPDM and PVC is poor, while the compatibility of CPE with PVC is better and related to the chlorine content of the CPE.

The impact strengths of PVC/EPDM and PVC/CPE at room temperature and $-12 \pm 2^\circ\text{C}$ are found in Figures 3 and 4 respectively. The effect of EPDM on PVC is insignificant due to their poor compatibility, even though EPDM is a good impact modifier in many other cases. Substantial impact strength modification is observed for the PVC/CPE polyblends at room temperature. At -12°C , the impact strengths of all blends are found to be 1/10 to 1/6 of the values at room temperature. Modification is also less significant. Impact strength modifications are affected by many factors such as the flex and glass transition temperatures and mechanical properties of the modifier, as well as compatibility between plastic and rubbery phases. Data indicates the best improvement can be obtained with CPE36.

Tricomponent Polyblends (PVC/CPE/EPDM)

Figure 5(a) illustrates the typical morphology of the PVC/CPE/EPDM polyblend. The dark regions are the EPDM phase, the gray area CPE phase,

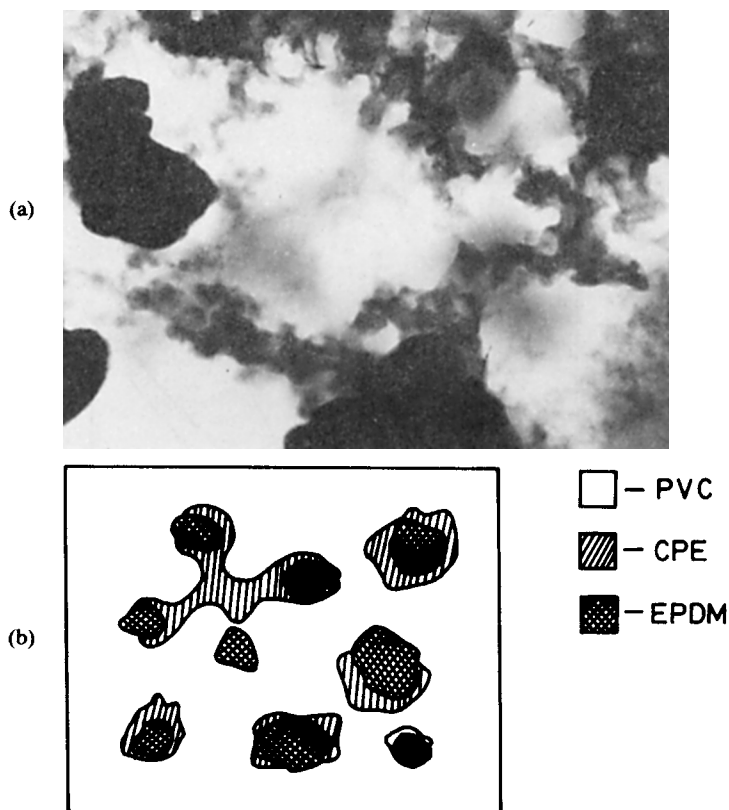


Fig. 5. Morphology of PVC/CPE36/EPDM (100/10/10; PHR) tricomponent polyblend. (a) Observed by TEM, $24,000 \times$. (b) Illustrated structural model.

TABLE III
Mechanical Properties of PVC, PVC/EPDM
and PVC/CPE/EPDM Polyblends

Property Polyblend	Yield T.S. (Kgf/cm ²)	Yield elong. (%)	Break elong. (%)	Modulus (Kgf/cm ²)
PVC	570 ± 35	6.71 ± 0.2	11.7 ± 0.4	12600 ± 350
PVC/EPDM 100/10 (PHR)	350 ± 27	5.90 ± 0.1	7.0 ± 0.2	11100 ± 260
PVC/CPE25/EPDM 100/10/10 (PHR)	340 ± 25	6.20 ± 0.3	19.6 ± 0.5	8900 ± 180
PVC/CPE25/EPDM 100/20/10 (PHR)	270 ± 18	6.60 ± 0.5	92.6 ± 1.3	7300 ± 195
PVC/CPE36/EPDM 100/10/10 (PHR)	340 ± 20	6.30 ± 0.3	24.1 ± 0.8	9000 ± 235
PVC/CPE36/EPDM 100/20/10 (PHR)	310 ± 15	6.5 ± 0.4	33.8 ± 0.8	8100 ± 150
PVC/CPE42/EPDM 100/10/10 (PHR)	360 ± 27	6.60 ± 0.2	13.8 ± 0.4	9400 ± 195
PVC/CPE42/EPDM 100/20/10 (PHR)	300 ± 15	6.60 ± 0.2	50.1 ± 1.1	7400 ± 150

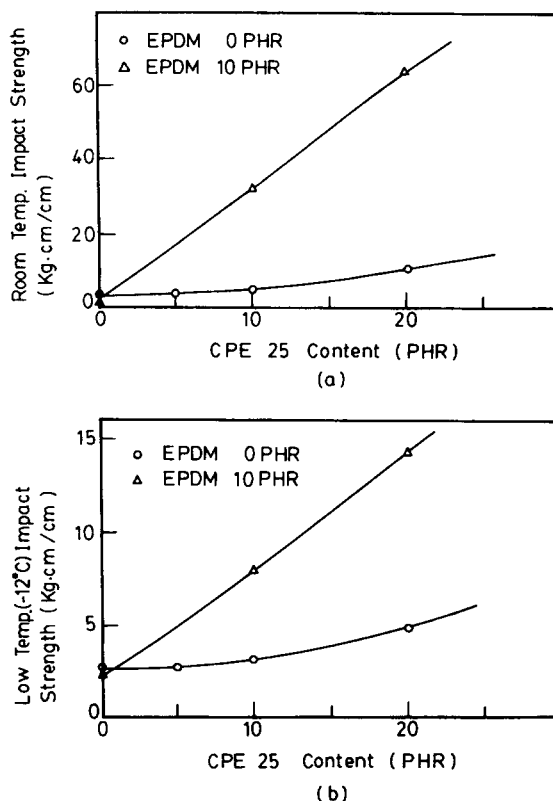


Fig. 6. Notched impact strengths of PVC/CPE25/EPDM polyblend. (a) Measured at room temperature. (b) Measured at -12°C .

and the white portions the PVC matrix. Well-defined boundaries exist between EPDM and PVC. This is expected because the interfacial adhesion between the two polymers is poor as observed in other polyblends such as PVC/PB (polybutadiene).^{13,14} However, a less distinct interface and, therefore, better compatibility is found between PVC and CPE, as well as CPE and EPDM. A similar phenomenon has been observed in the PVC/NBR (acrylonitrile butadiene rubber) polyblend.^{13,14} Therefore, location of CPE between PVC and EPDM is able to improve compatibility between the two, just as the effect of CPE on PVC/PE polyblends.^{4,6}

Based on TEM studies, a structural model for the PVC/CPE/EPDM is proposed, which, as illustrated in Figure 5(b), has the following features: (i) EPDM domains are dispersed in the PVC matrix. (ii) CPE phase is either encapsulating the EPDM domains or simply dispersed in the PVC matrix. (iii) Fraction of EPDM domains covered is proportional to the CPE/EPDM ratio.

Three effects are the direct results of this model. At high temperature, CPE reduces the yield stress and facilitates cold drawing of PVC,^{15,16} thus enhancing impact strength. At low temperature, CPE is a glassy material and poor impact modifier. However, EPDM has excellent impact resistance at low temperature. EPDM domains encapsulated by CPE, are able to induce cold

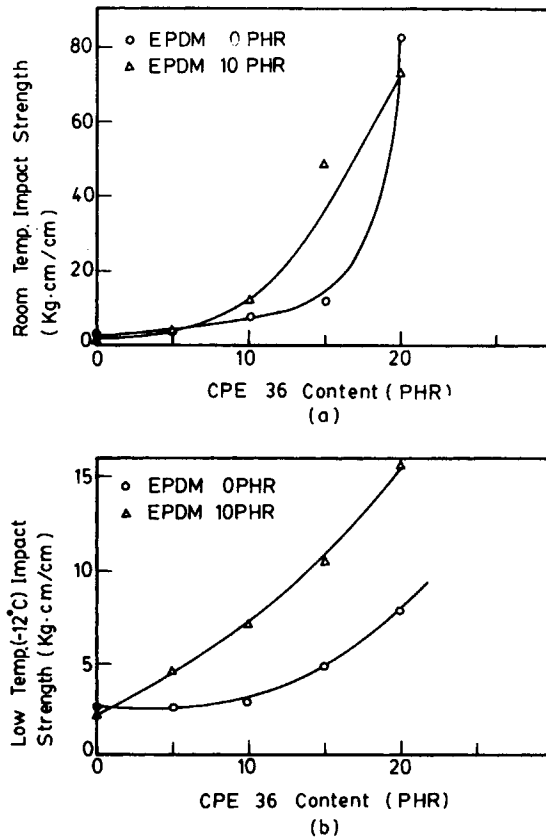


Fig. 7. Notched impact strengths of PVC/CPE36/EPDM polyblend. (a) Measured at room temperature. (b) Measured at -12°C .

drawing of CPE and, in turn, toughen the whole polyblends. Moreover, due to the poor adhesion between PVC and unencapsulated EPDM, cracks may be initiated at the interface, reducing the impact strength of the polyblends. The observed mechanical behavior of PVC/CPE/EPDM polyblends can be explained by the combination of these contributing effects.

Table III lists mechanical properties of PVC/CPE/EPDM polyblends. EPDM reduces the yield tensile strength, yield elongation, and modulus of PVC. Addition of CPE further reduces the yield tensile strength and modulus. The yield elongation increases slightly because of better adhesion caused by CPE encapsulation of EPDM.

Data in Figures 6, 7, and 8 demonstrate that the addition of 10 PHR EPDM to PVC/CPE polyblends results in an increase in impact strength at both room and low temperatures, except when CPE36 content is greater than 20 PHR at room temperature. Figure 9 illustrates the effect of EPDM content on the strength of the tricomponent PVC(100 PHR)/CPE36(20 PHR)/EPDM polyblends. Due to the glassy nature of CPE, the impact strength is low. If only a small amount of EPDM is added, most of the EPDM will be encapsulated. The impact toughening due to encapsulation is greater than the ruinous effect of unencapsulated EPDM. As EPDM content increases, these compet-

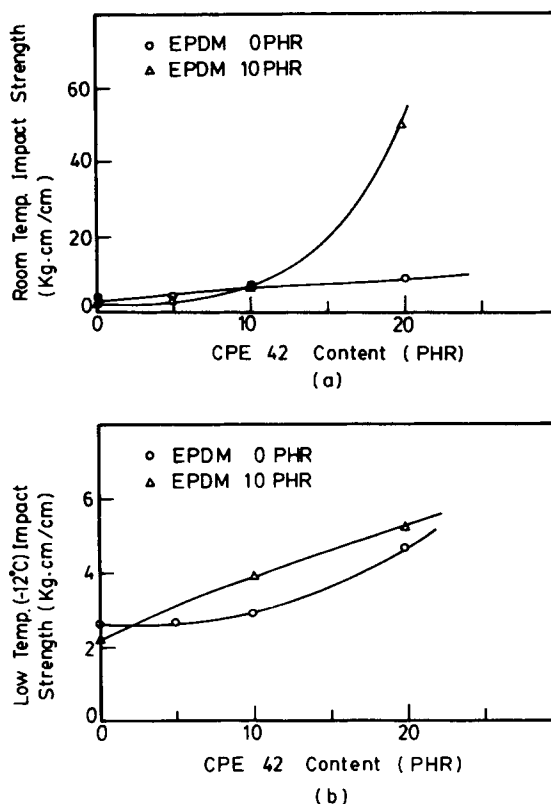


Fig. 8. Notched impact strengths of PVC/CPE42/EPDM polyblend. (a) Measured at room temperature. (b) Measured at -12°C .

ing effects produce a maximum in impact strength enhancement. At room temperature, since CPE36 is already a good impact modifier, encapsulation of added EPDM has a much smaller influence on the impact-toughening effect of CPE36. Exposed EPDM, however, produces interfacial cracks, reducing the impact strength. Thus, maximum enhancement is found at a much lower content. Other types of polyblends are proved to be poorer impact modifiers, and addition of encapsulated EPDM will substantially increase their impact-toughening ability. Impact strength reduction with addition of EPDM due to poor adhesion of exposed EPDM with the matrix is not observed in the experimental range.

CONCLUSION

CPE is a good impact modifier for PVC at room temperature, but becomes less so at low temperature due to its glassy nature. Best improvement is obtained with CPE36, since it has the lowest flex temperature and good compatibility with PVC. Addition of a suitable amount of EPDM will enhance impact toughening, however, too much EPDM reduces the impact strength of the polyblend. The experimental observation of impact-toughening

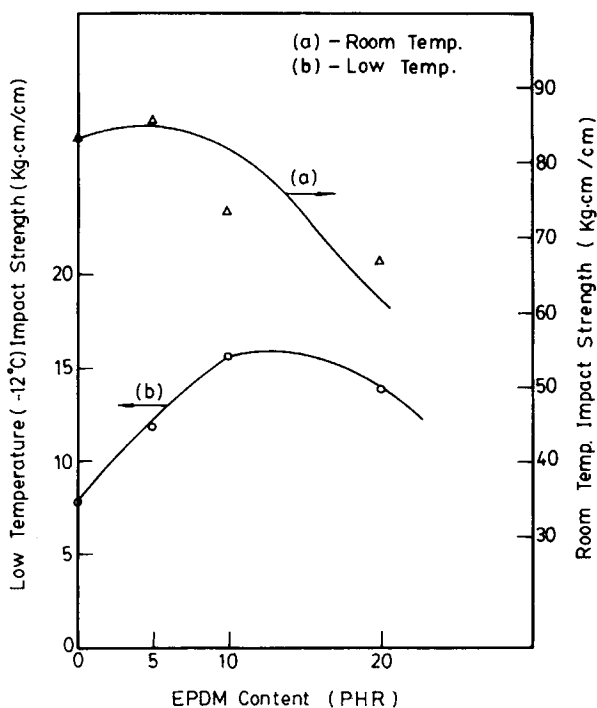


Fig. 9. Effect of EPDM content on the notched impact strengths of PVC/CPE36/EPDM polyblend. PVC content: 100 PHR, CPE36 Content: 20 PHR. (a) Measured at room temperature. (b) Measured at -12°C .

behavior can be explained by different toughening and ruinous mechanisms in a structural model.

References

1. A. Takahashi, *Polym. Eng. and Sci.*, **22**(1), 48 (1982).
2. T. O. Purcell, Jr. and R. E. Devaleria, *Plastics Engineering*, April, 46 (1976).
3. M. Matsuo, A. Ueda, and Y. Kondo, *Polym. Eng. Sci.*, **10**(5), 253 (1970).
4. C. E. Locke and D. R. Paul, *Polym. Eng. Sci.* **13**(4), 308 (1973).
5. D. R. Paul, C. E. Vinson, and C. E. Locke, *Polym. Eng. Sci.*, **12**(2), 157 (1972).
6. D. R. Paul, C. E. Locke, and C. E. Vinson, *Polym. Eng. Sci.*, **13**(3), 2092 (1973).
7. J. A. Brydson, *Rubber Chemistry*, Applied Science Publishers Ltd., London, (1978) Chap. 12.
8. Maurice Morton, *Rubber Technology*, Litton Education Publishing, Inc., (1973), Chap. 9.
9. D. R. Paul and S. Newman, *Polymer Blends*, Academic Press, New York, (1978), Vol. 2, p. 350.
10. R. R. Durst, R. M. Griffith, A. J. Urbanic, and W. J. Van Essen, *Amer. Chem. Soc. Adv. Chem. Ser.*, **154**, 239 (1976).
11. H. J. Oswald and E. T. Kubu, *SPE Trans*, **3**, 168 (1963).
12. D. Fleisher, E. Fischer, and J. Brandrup, *J. Macromol. Sci. Phys.*, **B14**(1), 17 (1977).
13. J. A. Manson and L. H. Sperling, *Polymer Blends and Composites*, Plenum Press, New York, (1976), Chap. 3, p. 89.
14. M. Matsuo, *Japan Plastics*, **2**, 6 (1968).
15. S. Newman and S. Strella, *J. Appl. Polym. Sci.*, **9**, 2297 (1965).
16. R. P. Petrich, *Polym. Eng. Sci.*, **13**, 248 (1973).

Received June 10, 1986

Accepted June 20, 1986