

Influence of interaction promoter on the properties of thermoplastic elastomeric blends of natural rubber and polyethylene

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The influence of a third component as interaction promoter on the properties of natural rubber-polyethylene thermoplastic blends, both uncured and cured, has been studied. The third component chosen has some structural similarity with polyethylene and is amorphous in nature. Ethylene propylene diene (EPDM) rubber, chlorinated polyethylene and chloro-sulphonated polyethylene have been used as the third component. All the third components have better adhesion with the plastic phase and the rubber phase. The adhesive strength is highest with EPDM. The properties are improved by using the above third components both for cured and uncured blends. In comparing the properties, the strength of the composite is divided by the modulus of the composite to take care of the hard-phase contribution. The size of the dispersed domain is reduced by using the third component and is approximately $1.2 \mu\text{m}$. All the properties could be explained in terms of the strengths of the individual phases, the morphology and the adhesion between components.

1. Introduction

In recent years, elastomeric rubber-plastic blends have become technologically important for many applications. They have many of the properties of rubbers, yet they can be processed as thermoplastics and do not need vulcanization. They offer a substantial economic advantage in respect of the fabrication into end-use parts. Although a large number of rubber-plastic blends are theoretically possible, only few of them have technological applications, due mainly to incompatibility and processing difficulties. Various rubber-plastic blends have been discussed by Coran [1], Kresge [2] and Morris [3].

The properties of a blend, such as modulus or strength, are a function of the properties of the blend components, phase morphology and the adhesion between the components. For example, molecular interdiffusion may increase the wetting of one phase by the other and reduce the effective interfacial tension. Increased adhesion would be expected to confer improved properties on the blends.

Though most of the rubber-plastic blends are technologically compatible at the melting temperature, they show segregation into respective phases upon cooling. If there is a great difference in hardness or shear modulus of the rubber and plastic (for example, highly crystalline plastics will display higher hardness and a larger difference in hardness values between rubber and plastic will result), there will be a sharp stress-gradient at the interface upon the application of force. The stress-transfer will not be uniform at the interface. In order to reduce this stress-gradient arising out of the incompatibility of the blend components, several investigators [4, 5] have reported the

technological enhancement of compatibility. Paul [6] studied the effect of block copolymers as interfacial agent to attain compatibility in a ternary blend system. Most of the studies so far made on ternary blends are based on the styrene-butadiene or styrene-isoprene block co-polymers. Molau and Wittbrodt [7] investigated the morphology of these ternary blends. According to their views, the incorporation of the third component reduces the particle size, thereby increasing interfacial adhesion and resisting the resultant gross phase segregation. The third component hence acts as an interaction promoter between the various phases.

Natural rubber-based thermoplastic elastomers are very promising in natural rubber-producing countries. Most of the studies concentrated in this field are on natural rubber-polyethylene and natural rubber-polypropylene blends [8, 9]. In the present investigation, natural rubber-polyethylene blends have been chosen and the interaction promoter has been selected in such a way that it has some structural similarity with the plastic phase and is rubbery in behaviour. The expectation is to have better interaction between the components under normal condition of application. Ethylenpropylene diene (EPDM) rubber, chlorinated polyethylene (CPE) and chlorosulphonated polyethylene (CSPE) have been used as the interaction promoter.

2. Experimental procedure

2.1. Materials used

Natural rubber (ISNR 5) was supplied by the Rubber Board, Kottayam, [specific gravity $\rho = 0.92$, solubility parameter $\delta = 8.1 \text{ cal}^{1/2} \text{ cm}^{-3/2}$ ($1 \text{ cal} = 4.19 \text{ J}$)].

TABLE I Formulations of the uncured mixes

Composition (parts by weight)	70/0/30	70/10/30	70/20/30	50/0/50	50/10/50	50/20/50	30/0/70	30/10/70	30/20/70	90/30
<i>Components</i>										
NR/EPDM/PE	A ₀	A ₁₀	A ₂₀	B ₀	B ₁₀	B ₂₀	C ₀	C ₁₀	C ₂₀	R ₀
NR/CPE/PE	A ₀	A' ₁₀	A' ₂₀	B ₀	B' ₁₀	B' ₂₀	C ₀	C' ₁₀	C' ₂₀	-
NR/CSPE/PE	A ₀	A'' ₁₀	A'' ₂₀	B ₀	B'' ₁₀	B'' ₂₀	C ₀	C'' ₁₀	C'' ₂₀	-
NR/EPDM/HAF*/PE	A _{B0}	-	A _{B20}	-	-	-	-	-	-	-
NR/CPE/HAF*/PE	-	-	A _{B20}	-	-	-	-	-	-	-
NR/CSPE/HAF*/PE	-	-	A _{B20}	-	-	-	-	-	-	-

*30 parts of HAF black was added to the rubber phase.

Polyethylene (Indothene 16 MA 400) was supplied by IPCL, Baroda ($\rho = 0.916$, $\delta = 7.9$). Chlorosulphonated polyethylene was supplied by Du Pont, USA ($\rho = 1.18$, $\delta = 9.0$). Chlorinated polyethylene (36% chlorine) was supplied by Dow Chemicals ($\rho = 1.16$, $\delta = 9.0$). Ethylene propylene diene rubber (Keltan 520) was supplied by DSM, Holland ($\rho = 0.86$, $\delta = 7.95$).

2.2. Blend preparation

Binary and ternary blends were prepared by melt-mixing the polymers in a Brabender Plasticorder (Model PLE 330) fitted with a cam-type mixer with a rotor speed of 60 r.p.m., residence time of 6 min and mixer chamber temperature set at 150°C. Generally, the plastic was melted in the mixer for 1 to 2 min; the masticated rubber strips were then added and allowed to blend for 4 min. After blend formation, curatives were added wherever required and the mixing continued until the torque increased by 2 to 3 units, indicating the progress of cross-linking.

A few black composites were also made. First a rubber-black master-batch was prepared, and from this a definite proportion was taken in a strip form and allowed to blend with the molten plastic in the Brabender mixer under similar conditions.

The blend was taken out from the mixer and passed through a laboratory mill set at 2.0 mm nip setting to get a sheet. The compositions of the investigated blends are reported in Tables I and II.

The premixed material was compression-moulded between aluminium foils at 150°C for 3 min at a pressure of 0.25 tons per sq. in. (3.9 MPa) in an electrically heated press to get slabs of 14 cm × 14 cm × 0.2 cm size. The aluminium foils were used to reduce shrink-marks in the mouldings. After completing the moulding, the sample (still under compression) was immediately cooled in water to avoid the possible degradation

of the rubber phase under high-temperature exposure and to maintain the overall dimensional stability of the blend.

2.3. Preparation of samples for adhesion

In order to prepare samples for adhesion, the technique adopted consisted of two steps. Thin sheets of rubber of 2 mm thickness were made by the moulding technique using a gentle pressure and at 100°C. One side of the rubber was fabric-backed. The plastic was also used in a sheet form.

Then the plastic sheet was put over the fabric-backed rubber sheet, in between which cellophane paper was partially introduced to get a demarcation line for gripping during testing. The assembly was then heated for 10 min at 100°C followed by 15 min at 150°C.

2.4. Measurement of technical properties

The mechanical properties of the blends were determined according to the ASTM D 412-80 test method, using dumb-bell-shaped test pieces. The test pieces were punched out from the moulded sheets by a hollow punch, along the mill-grain direction of the sheets.

The stress-strain curves in tensile mode at room temperature were obtained by means of a Zwick UTM apparatus (Model 1445) at a crosshead speed of 200 mm min⁻¹.

Hysteresis tests were also performed on the dumb-bell specimen at room temperature in the Zwick UTM. One cycle between two definite force intervals was selected. The testing rate for both loading and unloading was 50 mm min⁻¹. The hysteresis loss (W_2) during the complete cycle was calculated and the total strain energy was also found out and represented by (W_1).

2.5. Electron microscopy studies

Blends were characterized with the help of a JEOL transmission electron microscope and a Phillips 500 scanning electron microscope. Some of the samples were solvent-extracted for 2 days at room temperature using n-hexane, whereby the rubbery phase was removed from the blend. Dynamically cross-linked samples were etched with nitric acid for 2 days at room temperature.

All the samples for SEM studies were sputter-coated with gold and examined within 24 h of testing.

TABLE II Formulation of the cured mixes

Component	Composition (parts by weight)				
	A _{c0}	A _{c20}	A' _{c20}	A'' _{c20}	R _{c0}
NR	70	70	70	70	90
PE	30	30	30	30	30
EPDM	-	20	-	-	-
CPE	-	-	20	-	-
DCP	0.5	0.5	0.5	0.5	0.5
CSPE	-	-	-	20	-
HAF black	-	-	-	-	-

A fractographic analysis of the ruptured surfaces was also carried out using SEM.

2.6. Measurement of surface energy of blends

The surface energy of various rubbers and plastics was determined using a contact angle meter (Kernco Model GII). The method is based on the measurement of contact angles by the sessile drop method with water and glycerol.

According to Fowkes [10] the total free energy at a surface is the sum of contributions from the different intermolecular forces at the surface. The following relation given by Fowkes for the contact angle of a liquid on a solid surface was used:

$$1 + \cos \theta = 2(\gamma_s^d)^{1/2} \frac{(\gamma_l^d)^{1/2}}{\gamma_{lv}} + 2(\gamma_s^h)^{1/2} \frac{(\gamma_l^h)^{1/2}}{\gamma_{lv}} \quad (1)$$

where θ = angle of contact between the solid and liquid surface; γ_s^h, γ_s^d = components of surface energy of solid due to hydrogen bonding and dipole-dipole interaction, respectively; γ_{lv} = surface free energy of liquid; and γ_l^h, γ_l^d = components of surface energy of liquid due to hydrogen bonding and dispersion force component, respectively.

2.7. Measurement of strength of adhesion

180° peel tests were performed for measuring the adhesive strength. The strength was calculated using the formula

$$G_a = 2F/w \quad (2)$$

where G_a = adhesive strength, F = peeling force and w = width of the sample. All the adhesion measurements were carried out at room temperature and at 200 mm min⁻¹ rate.

3. Results and discussion

3.1. Phase morphology of the blends

3.1.1. Uncured blends

The blend morphology of the NR/PE = 70/30 system (Blend A₀) is shown in Fig. 1. The morphology study was carried out after removing the surface rubber by n-hexane. The size of the rubber particles is not uniform, as shown in Fig. 2 at high magnification. With the introduction of EPDM or CPE, the size of the

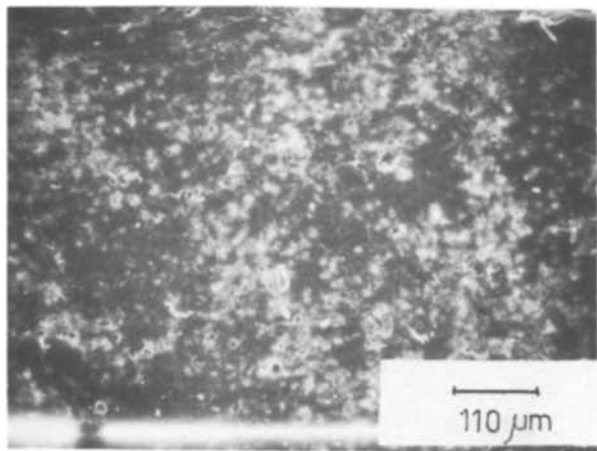


Figure 1 SEM photograph of extracted surface of Blend A₀.

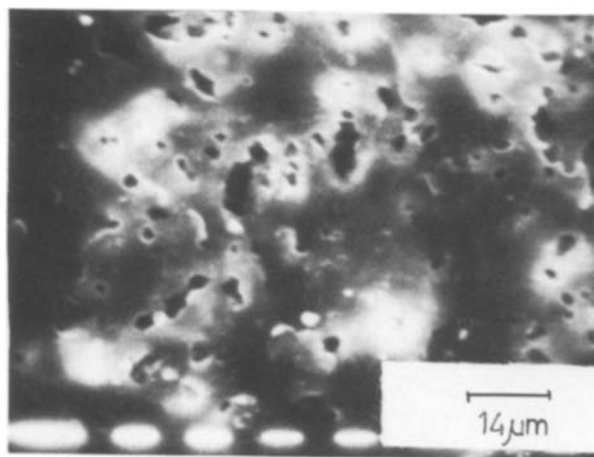


Figure 2 Magnified view of the extracted surface of Blend A₀.

dispersed particles is reduced by 33% (Fig. 3). The average diameter of the dispersed rubber domain in NR/EPDM/PE is 1.2 μm and both phases are continuous. The low viscosity of the plastic at the mixing temperature accompanying the high proportion of rubber in the mixes studied favours the continuity of both the phases. EPDM, CPE or CSPE, having some structural similarities with polyethylene and amorphous characteristics like rubber, reduce the particle size because of better adhesion [1].

3.1.2. Cured blends

On curing the rubber phase, we observe that rubber and plastic remain co-continuous (Fig. 4). The third component reduces the size of the rubber particles (Fig. 5). In order to determine the size of the particle correctly, TEM of the blends was carried out. The information obtained from SEM after extracting with the solvent and nitric acid etching is in line with that obtained from TEM after staining the samples with osmium tetroxide.

3.2. Properties of unvulcanized rubber-plastic blends

The properties of various natural rubber-polyethylene blends are shown in Tables III to V. As pointed out before, EPDM, CPE and CSPE used as a third component has been added at a level of 10 and 20 p.h.r. (parts per hundred of rubber-plastic).

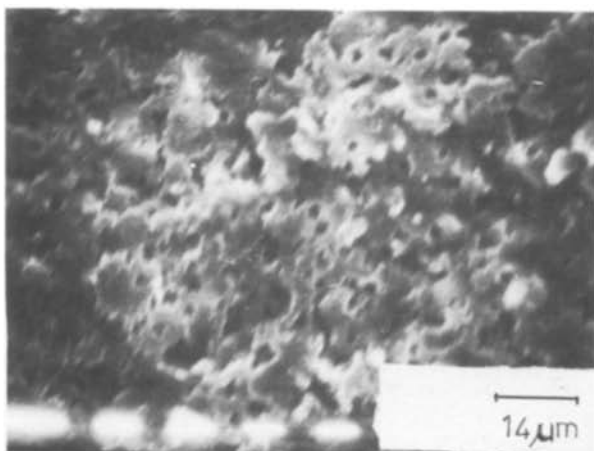


Figure 3 SEM photograph of extracted surface of blend A'₂₀.

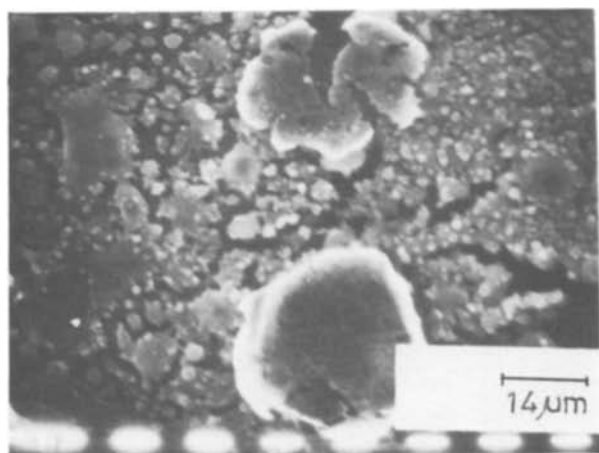


Figure 4 SEM photograph of etched surface of blend A_{c_0} .

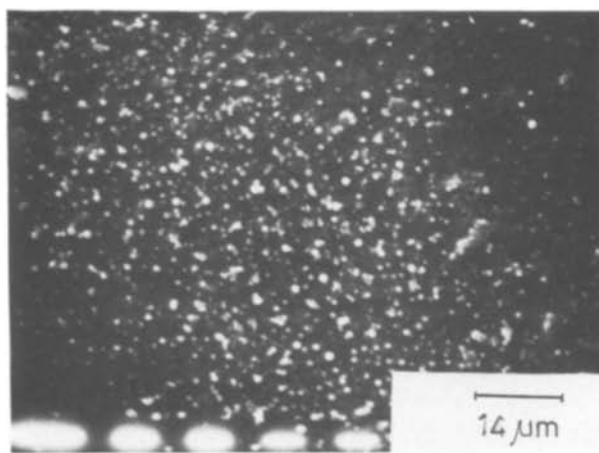


Figure 5 SEM photograph of etched surface of blend $A_{c_{20}}$.

With the introduction of EPDM into NR/PE blends, the modulus at 100% elongation decreases. The tensile strength of 70/30 NR/PE blends is reduced, while 50/50 and 30/70 blends show an optimum in strength with 10 p.h.r. of EPDM. The elongation at break, however, increases with the increase in EPDM content. When compared with the properties of the rubbers and plastics, the tensile strength and modulus are close to those of rubber. The elongation at break for 70/30 NR/PE blends, however, is a little higher.

With the introduction of CPE or CSPE, the strength increases or remains constant, though the modulus at 100% elongation decreases. For example, in the case of the 70/20/30 blend using CPE there is an increase of 21% in strength over the control sample. Similarly the strength of 30/10/70 blends using 10 p.h.r CSPE is 5.49 (an increase of 32%). However, when all the blends are compared with R_0 , a 90/30 NR/PE blend in which the third component is replaced by 20 parts of NR, the tensile strength and the modulus consistently increase. Sometimes the enhancement is 1.5 times.

In all these cases, when the plastic component is increased, (c.f. 70/30 and 30/70), the blends show enhanced strength due to the higher strengths of the plastic phase. Hence, a correction is made by dividing the strength by the 100% modulus and by the strength of the hard phase. The results are reported in Tables III to V. For a particular blend system, the ratio σ_b/E increases with the incorporation of the third component. This is very prominent in 70/30 blends. This means that the third component, which has a similarity in part of its structure with the plastic phase, actually increases the strength of the blends. The reason for this enhancement is discussed later. Similarly, σ_b/σ_H has been calculated for all the blends. This remains constant or increases marginally.

The hysteresis properties of various blends are reported in Table VI. The strain energy (W_1) and the hysteresis loss (W_2) have been calculated, at forces between 0.001 and 15 N for one cycle. NR/CSPE/PE systems show more hysteresis loss as compared to the NR/PE system at all compositions (70/30, 50/50, 30/70). However, for NR/EPDM/PE and NR/CPE/PE systems, the above trend is followed

TABLE III Properties of the uncured NR/EPDM/PE blends

Property	Blend									
	A_0	A_{10}	A_{20}	B_0	B_{10}	B_{20}	C_0	C_{10}	C_{20}	R_0
100% modulus, E (MPa)	1.69	1.56	1.39	3.47	3.04	2.55	—	—	—	0.95
Tensile strength, σ_b (MPa)	3.6	3.46	3.40	3.81	4.23	3.18	4.16	5.13	4.36	2.65
Elongation at break (%)	560	440	600	180	340	310	7	45	65	510
σ_b/E	2.1	2.21	2.44	1.09	1.39	1.25	—	—	—	2.78
σ_b/σ_H^*	0.36	0.36	0.35	0.39	0.43	0.33	0.43	0.53	0.45	0.27

*Tensile strength of the hard phase.

TABLE IV Properties of the uncured NR/CPE/PE blends

Property	Blend									
	A_0	A'_{10}	A'_{20}	B_0	B'_{10}	B'_{20}	C_0	C'_{10}	C'_{20}	R_0
100% modulus, E (MPa)	1.69	1.38	1.57	3.47	—	3.3	—	—	—	0.95
Tensile strength, σ_b (MPa)	3.6	4.0	4.61	3.81	3.07	3.39	4.16	4.36	4.05	2.65
Elongation at break (%)	560	480	490	180	142	145	7	10	15	510
σ_b/E	2.1	2.89	2.93	1.09	—	1.03	—	—	—	2.78
σ_b/σ_H^*	0.36	0.41	0.47	0.39	0.32	0.35	0.43	0.45	0.42	0.27

*Tensile strength of the hard phase.

TABLE V Properties of the uncured NR/CSPE/PE blends

Property	Blend									
	A ₀	A'10	A'20	B ₀	B'10	B'20	C ₀	C'10	C'20	R ₀
100% modulus, <i>E</i> (MPa)	1.69	1.18	0.97	3.47	3.34	2.71	–	–	–	0.95
Tensile strength, σ _b (MPa)	3.6	3.35	3.78	3.81	4.17	3.51	4.16	5.49	5.3	2.65
Elongation at break (%)	560	485	565	180	240	305	7	25	40	510
σ _b / <i>E</i>	2.1	2.85	3.98	1.09	1.25	1.29	–	–	–	2.78
σ _b /σ _H *	0.36	0.36	0.39	0.39	0.43	0.36	0.43	0.56	0.54	0.27

*Tensile strength of the hard phase.

TABLE VI Hysteresis (uncured systems)

	A ₀	A ₁₀	A ₂₀	B ₀	B ₁₀	B ₂₀	C ₀	C ₁₀	C ₂₀	R ₀
NR/EPDM/PE										
W ₂ (Nm) × 10 ²	141.5	*	*	6.8	4.4	26.8	0.6	0.6	1.0	172
W ₁ (Nm) × 10 ²	158.4	*	*	9.5	6.9	34.6	1.4	1.4	2.2	200
	A ₀	A'10	A'20	B ₀	B'10	B'20	C ₀	C'10	C'20	R ₀
NR/CPE/PE										
W ₂ (Nm) × 10 ²	141.4	140.1	107.1	6.8	3.7	4.8	0.6	0.7	0.9	172
W ₁ (Nm) × 10 ²	158.5	161.8	128.6	9.5	5.9	7.4	1.4	1.5	1.8	200
	A ₀	A'10	A'20	B ₀	B'10	B'20	C ₀	C'10	C'20	R ₀
NR/CSPE/PE										
W ₂ (Nm) × 10 ²	141.5	148.9	167.9	6.8	8.9	138.8	0.6	0.5	0.9	172
W ₁ (Nm) × 10 ²	158.4	171.1	196.3	9.5	12.0	152.2	1.4	1.4	1.9	200

*Samples failed under the testing condition.

only for 30/70 blends. 70/30 and 50/50 blends do not show any particular trend. When the proportion of the plastic phase is more, it is understandable that the hysteresis loss will be less. It is apparent from our data that the EPDM-containing blends display low hysteresis losses. As discussed later, this may be related to the higher adhesion of EPDM to both NR and PE. In the case of CSPE-containing blends, the higher hysteresis may be related to higher elongation in the force range used. NR/CPE/PE shows intermediate behaviour.

3.3. Properties of the blends prepared by dynamic vulcanization

Since the green strength of EPDM, NR and CSPE is expected to be low, a few parts of dicumyl peroxide were added in the mix for dynamic vulcanization. The properties of the various mixes are reported in Table VII. With the addition of peroxide, the strength value of the blends is almost doubled. The NR/CSPE/PE mix has the lowest modulus, tensile strength and elongation at break, but the highest hysteresis loss. The mix containing CPE is best in the series in tensile strength. When a correction is made for the hard-phase component, NR/CPE/PE is comparable to the NR/PE system. However, in the NR/CPE/PE system, the rubber-to-plastic phase ratio is 3:1 for 70/20/30 blends. Hence, a mix containing 90/30 NR/PE (mix R_{c0}, Table II) was compared with the above system. It was observed that the strength, modulus, σ_b/*E* and σ_b/σ_H were increased. It could be concluded, as discussed later, that the third phase used in the present system actually enhances the strength properties.

3.4. Fracture surface analysis

The fracture surfaces of various systems in uncured blends do not show any distinct differences. As shown in Fig. 6, a rough fracture surface with a few flow lines is generated. On curing, the surface shows many parabolic lines and flow deviations indicative of high-strength materials (Fig. 7). Again, there is not much difference among the various blends studied.

3.5. Adhesion between the components and correlation between adhesion and strength properties

The surface energy γ_s of the various rubbers and plastics is shown in Table VIII. The γ_s values of NR and PE are similar. Though the value of γ_s for EPDM is close to that of NR, or PE, that of CPE is widely different.

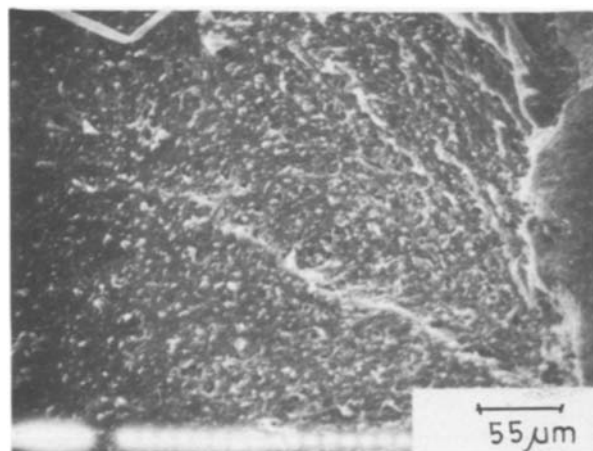


Figure 6 Tensile fractograph of blend A₂₀.

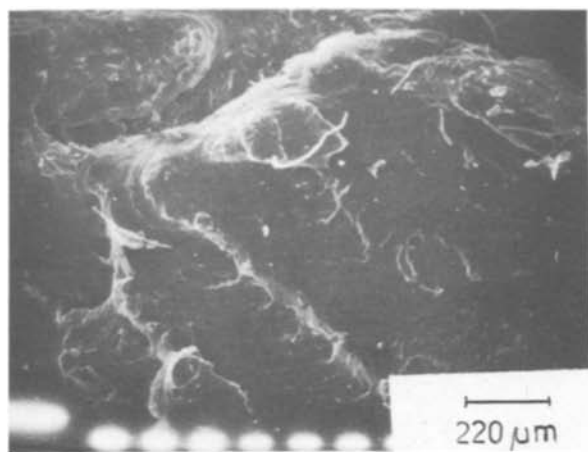


Figure 7 Tensile fractograph of blend A_{c20} .

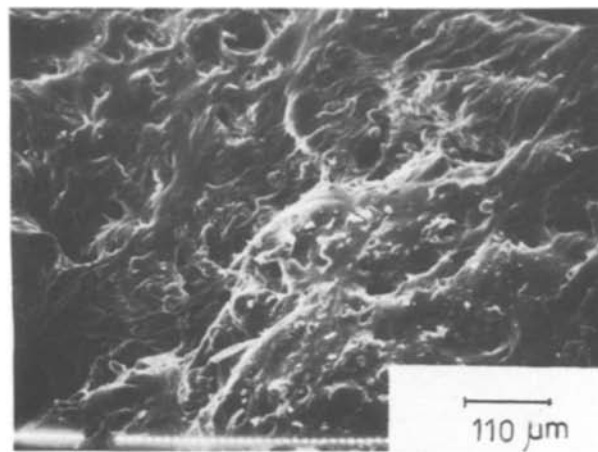


Figure 8 SEM photograph of peeled NR/EPDM surface.

Table IX indicates the values of peel adhesion. Natural rubber–polyethylene shows a value of 140 J m^{-2} . With the addition of EPDM to the NR phase the value increases to 3190 J m^{-2} , because the adhesive strengths of NR/EPDM (1330 J m^{-2}) and EPDM/PE (1440 J m^{-2}) are higher. The composite shows a synergistic effect. This may be explained in terms of a similar value of γ_s . However, with CPE, when the γ_s value is different, the peel strength increases marginally. A similar increase is observed in the case of CSPE. This is reflected in the adhesive strength of NR/CPE and CPE/PE or NR/CSPE and CSPE/PE. These values are higher than for NR/PE systems, which may be due either to interdiffusion between the amorphous phases and the amorphous/crystalline phases, or partial structural similarity. Cross-linking of the rubber phase increases the strength of adhesion quite significantly. For example, there is a 43% increase for NR/EPDM/PE and 66% for NR/CPE/PE systems over the control samples.

This is due to the increase in strength of the weaker substrate.

The enhancement in adhesive strength due to addition of the third component is also observed on the peeled surface. For example, on addition of the third component, the peeled rubber surface shows cavities all over (Fig. 8). These are however absent on the surface of the control sample (Fig. 9). This also indicates that the EPDM phase promotes adhesion with its presence at the interface. In this sense, the third components used here are acting as interaction promoters.

It must be pointed out that the increase in properties is not concomitant with the increase in strength of adhesion, though the level of adhesion is increased by incorporating the third component. Though EPDM has the highest adhesion between NR or PE, the properties of the composites are not the best among the systems studied. We have also calculated the surface mismatch $\Delta\gamma_{SH}$ by taking the difference

TABLE VII Properties of the blends prepared by dynamic vulcanization

	A_{c0}	A_{c20}	A'_{c20}	A''_{c20}	R_{c0}
100% modulus, E (MPa)	2.06	2.9	2.33	1.77	1.95
Tensile strength, σ_b (MPa)	7.92	8.24	9.2	5.16	7.61
Elongation at break (%)	480	384	457	359	217
σ_b/E	3.84	2.84	3.94	2.92	3.9
σ_b/σ_H^*	0.81	0.85	0.95	0.52	0.78
W_2 (Nm) $\times 10^2$	32.3	17.4	22.1	53.5	12.0
W_1 (Nm) $\times 10^2$	43.7	29.8	36.2	71.6	19.0

*Tensile strength of the hard phase.

TABLE VIII Surface energy for various solids

Surface	Surface tension, γ_{s1} (dyn cm^{-1})*	Surface	Surface tension, γ_{s2} (dyn cm^{-1})*	Difference in surface tension, $\Delta\gamma_{SH}$ (dyn cm^{-1})*
NR	32.5	PE	33.1	0.06
EPDM	29.7	PE	33.1	3.4
CSPE	36.5	PE	33.1	3.4
CPE	37.2	PE	33.1	4.1
NR/EPDM [†]	30.2	PE	33.1	2.9
NR/CPE [†]	28.5	PE	33.1	4.6
NR/CSPE [†]	26.0	PE	33.1	7.1

*1 dyn $\text{cm}^{-1} = 10^{-3} \text{ N m}^{-1}$.

[†] 20 parts of the third component was added to NR.

TABLE IX 180° peel test adhesion data

Sample	Adhesive strength (J m ⁻²)	Sample	Adhesive strength (J m ⁻²)	Sample	Adhesive strength (J m ⁻²)
NR/PE	140	NR/CPE	260	NR/PE/DCP	190
NR/CPE*/PE	150	NR/EPDM	1330	NR/CPE*/PE/DCP	250
NR/EPDM*/PE	3190	NR/CSPE	170	NR/EPDM*/PE/DCP	4560
NR/CSPE*/PE	250	CSPE/PE	200		
		CPE/PE	420		
		EPDM/PE	1440		

*20 parts of the third component was added to NR.

between individual γ_s values (Table VIII). However, we could not get any relationship between σ_b/σ_H and $\Delta\gamma_{SH}$ of the form shown by Coran and Patel [11]. This means that morphology and the strength of individual components play an important role. The morphology, as pointed out before, is almost the same for all the ternary systems. Hence, the difference must lie with the properties of the components. Stress-strain data of the individual polymers are shown in Fig. 10. CPE/CSPE shows a higher strength than EPDM because of the more crystalline component in the former. The enhancement in properties in NR/CPE/PE is due to increased green strength, while in the case of NR/EPDM/PE it is due to increased adhesion.

It is the interplay of adhesion and the strengths of individual components that determines the properties of the composites.

In all the above mixes, addition of 20 p.h.r. EPDM dilutes the plastic phase and increases the concentration of the rubber phase. Hence, the decrease in overall properties of some of the mixes having a ternary component is due to a dilution effect. In order to understand this effect, a mix was prepared with 90/30 NR/PE (mix R_0 in Tables III, IV and V). 20 p.h.r. of the third component was replaced by 20 p.h.r. of NR. The decrease in tensile strength (26%) is more than that of NR/EPDM/PE. This shows the role played by the interaction promoter in determining the composite properties. A further decrease in strength of the composite is observed when carbon black is added to NR/EPDM (Table X). Though the strength of the uncured rubber phase is increased, the adhesion falls except for CPE. The crystallinity of CPE is probably

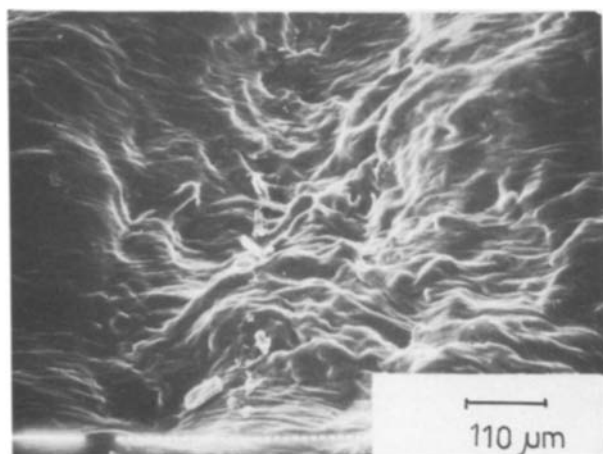


Figure 9 SEM photograph of peeled NR surface.

reduced by the addition of carbon black. It is also interesting to note from Table X that the blends $A_{B_{20}}$ and $A''_{B_{20}}$ show improved strength over A_{B_0} , when the strength of the substrate is enhanced. The adhesion values increase with the addition of the third component in the case of EPDM and CPE.

4. Conclusions

The effect of third components, namely EPDM, CPE and CSPE, on the properties of NR/PE blends has been studied. From the morphology it is evident that the third component reduces the domain size of the rubber in the NR/PE blends. These particles act as an alloying agent between NR and PE, indicating better interaction between the constituent polymers. Thus the role played by the third component in the NR/PE blend is nothing but an interaction promoter.

With the incorporation of the third component the modulus in all cases decreases due to lowering of the overall crystallinity. On the other hand the softening effect due to rubber addition gives rise to a higher elongation at break with some blend compositions, particularly at high plastic concentrations. However, the tensile strength increases with the addition of CPE but the effect is not so marked in cases of blends containing EPDM and CSPE. This trend is attributed to the green strength of the individual components. Furthermore, all the blends show higher strength at

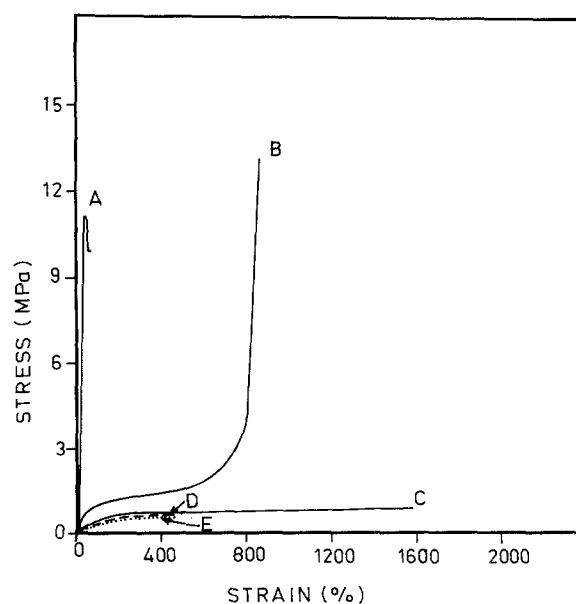


Figure 10 Stress-strain curves of the base polymers: (A) PE, (B) CPE, (C) CSPE, (D) NR, (E) EPDM.

TABLE X Properties of the uncured black composites

Property	Blend			
	A_{B_0}	$A_{B_{20}}$	$A'_{B_{20}}$	$A''_{B_{20}}$
100% modulus, E (MPa)	0.58	0.64	0.82	0.74
Tensile strength, σ_b (MPa)	1.15	1.75	0.83	1.41
Elongation at break (%)	224	225	270	280
σ_b/E	1.98	2.73	1.01	1.91
σ_b/σ_H^*	0.12	0.18	0.09	0.14
Adhesive strength (Jm^{-2})	224	614	384	130

*Tensile strength of the hard phase.

higher plastic concentration. Hence a correction is done by dividing the strength by the strength of hard phase and by the 100% modulus. This shows that the third component enhances the strength property.

This improvement is more prominent whenever we compare the 70/20/30 NR/third component/PE blends with the 90/30 NR/PE blend.

A similar trend is observed in case of dynamically vulcanised blends.

The NR/CSPE/PE blends show the best hysteresis property at all compositions, whereas EPDM-containing blends offer low hysteresis loss and NR/CPE/PE blends show an intermediate behaviour.

The level of adhesion is improved with the third component, and EPDM imparts the highest adhesive strength to the NR/PE blend. This can be explained by the small surface-energy mismatch. The other two blends (CPE and CSPE) also show improvements in the level of adhesion.

Thus, with the proper selection of the third component on the ground of surface chemical properties

and structural identity, a great change in the phase morphology with subsequent improvement in overall properties can be achieved.

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