

# Compatibility Studies on Some Polymer Blend Systems by Electrical and Mechanical Techniques

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**ABSTRACT:** Systematic electrical and mechanical studies were carried out on natural rubber (NR) blended with different types of synthetic rubber such as styrene-butadiene rubber (SBR), polybutadiene rubber (BR), and ethylene-propylene-diene monomer (EPDM) as nonpolar rubbers and nitrile-butadiene rubber (NBR) and chloroprene rubber (CR) as polar rubbers. The NR/SBR, NR/BR, NR/EPDM, NR/NBR, and NR/CR blends were prepared with different ratios (100/0, 75/25, 50/50, 25/75, and 0/100). The permittivity ( $\epsilon'$ ) and dielectric loss ( $\epsilon''$ ) of these blends were measured over a wide range of frequencies (100 Hz–100 kHz) and at room temperature ( $\sim 27^\circ\text{C}$ ). The compatibility results obtained from the dielectric measurements were comparable with those obtained from the calculation of the heat of mixing. These results were confirmed by scanning electron microscopy and showed that NR/SBR and NR/BR blends were compatible while NR/EPDM, NR/NBR, and NR/CR blends were incompatible. To overcome the problem of phase separation (incompatibility) between NR and EPDM, NBR, or CR, a third component such as SBR or poly(vinyl chloride) (PVC) was added as a compatibilizing agent to these blends. The experimental data of dielectric and mechanical measurements showed that the addition of either SBR or PVC could improve the compatibility of such blends to some extent. © 2000 John Wiley & Sons, Inc. *J Appl Polym Sci* 79: 60–71, 2001

**Key words:** compatibility studies; polymer blends; natural rubber; synthetic rubbers

## INTRODUCTION

The importance of the blending of two types of rubber has increased recently, because it is a useful approach for the preparation of new materials with desirable properties absent in the component rubber.<sup>1–4</sup> A particularly interesting example can be found in tire production. Natural rubber (NR) may be blended with synthetic rubber to improve building properties<sup>5</sup> (e.g., tensile strength, resilience, tear strength, fatigue fracture, etc.). The commercially useful polymer–polymer combination is linked by intermolecular

forces such as van der Waal's forces or dipole moments and exhibits sufficient thermodynamic compatibility to prevent the polymer phases from separating during melt processing.<sup>6</sup>

The blending of rubber with plastic to achieve good mechanical properties is not an easy process because of the incompatibility of most of these blends.<sup>7</sup> Very often the resulting materials exhibit poor mechanical properties due to the poor adhesion between the phases. Several trials were carried out to minimize the phase separation and increase interfacial adhesion; these included the addition of physical or chemical compatibilizers,<sup>8,9</sup> (the addition of a third homopolymer or graft or block copolymer) that bind with the two phases and the introduction of covalent bonds between the homopolymer phases.

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**Table I Specifications of Rubber Types**

Specification	Rubber					
	NR	BR	NBR	CR	EPDM	SBR
Specific gravity	0.913	0.915	1.170	1.230	0.860	0.945
Moony viscosity ML (1 + 4) at 100°C	41 ± 2	35 ± 3	45 ± 5	50 ± 5	85	52 ± 3
Ave. molecular weight <sup>a</sup>	174,189	140,326	163,376	379,711	—	140,326

<sup>a</sup> Calculated using the Huggins equation.<sup>12</sup>

The aim of the present work was to employ the dielectric measurements to investigate the compatibility of different types of rubber blends. Furthermore, the heat of mixing of polymer–polymer blends was calculated<sup>10</sup> and compared with the dielectric results in order to check the validity of the limiting value of the heat of mixing given<sup>11</sup> for compatible blends. The results obtained were further confirmed using scanning electron microscopy. This study was also concerned with the use of a third polymer as a compatibilizer for incom-

patible blends in order to change their compatibility behavior.

## EXPERIMENTAL

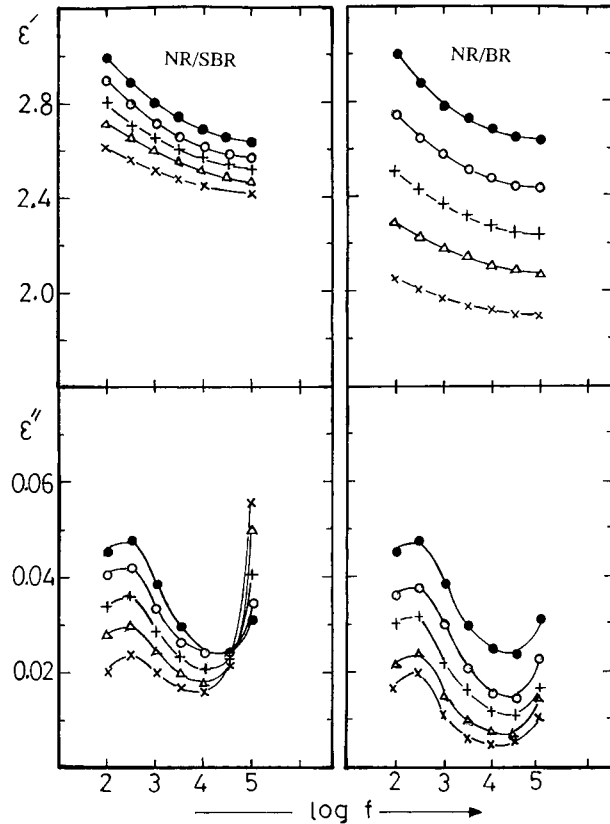
### Materials

The ribbed smoked sheets of NR (RSS-1) were supplied by Transport and Engineering Company (Alexandria, Egypt). The styrene-butadiene rub-

**Table II Rheometric Characteristics and Physicomechanical Properties of NR/SBR and NR/BR Blends**

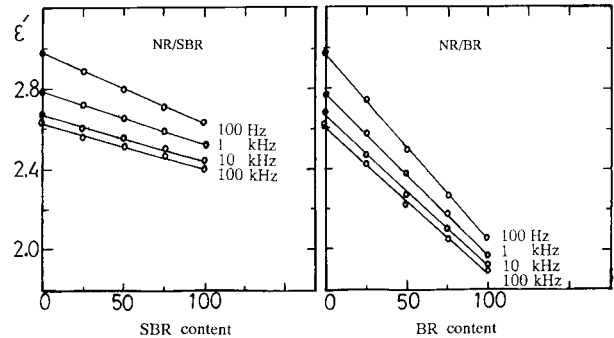
	Sample								
	A	A <sub>1</sub>	A <sub>2</sub>	A <sub>3</sub>	A <sub>4</sub>	B <sub>1</sub>	B <sub>2</sub>	B <sub>3</sub>	B <sub>4</sub>
Formulation									
NR	100	75	50	25	—	75	50	25	—
SBR	—	25	50	75	100	—	—	—	—
BR	—	—	—	—	—	25	50	75	100
Peroxide	4	4	4	4	4	4	4	4	4
Rheometric characteristics at 152 ± 1°C									
$M_L$ (dN m)	6.5	4.5	6	8.5	10	10.75	13.5	13	14
$M_H$ (dN m)	40	50	58	70.5	64	51	69	86.5	96.5
$t_{s2}$ (min)	2.5	3.0	3.25	2.75	2.25	2.75	2.13	1.5	1.75
$t_{c90}$ (min)	40.5	41.5	38.5	38.5	39.5	33	29.5	27.5	28.5
CRI (min <sup>-1</sup> )	2.63	2.59	2.84	2.79	2.68	3.3	3.65	3.5	3.73
Physicomechanical properties									
$M$ at 100% $E$									
(MPa)	0.36	0.52	0.63	0.71	0.78	0.47	0.607	0.67	—
TS (MPa)	1.014	0.99	0.98	0.94	0.8	1.03	0.8	0.69	0.59
Elongation (%)	225	185	165	158	125	200	150	125	90
Equilibrium swelling (%)									
	310.51	305	286	272	246	323	293.8	274.3	253.9
Soluble fraction (%)									
	3.53	4.2	4.91	5.43	6.54	2.85	2.72	2.43	2.05

The following definitions apply to Tables II–V:  $M_L$ , Minimum torque;  $M_H$ , maximum torque;  $T_{s2}$ , scorch time;  $T_{c90}$ , optimum cure time; CRI, cure rate index;  $M$  at 100%  $E$ , modulus at 100% elongation; TS, tensile strength.



**Figure 1** The permittivity ( $\epsilon'$ ) and dielectric loss ( $\epsilon''$ ) versus the frequency ( $\log f$ ) for NR/SBR and NR/BR rubber blends with (●) 100, (○) 75, (+) 50, ( $\Delta$ ) 25, and (×) 0 parts of NR in the blend.

ber (SBR1502) with 23.5% styrene content was also supplied by Transport and Engineering Company. The polybutadiene rubber (BR) was in the 1,4-cis form (97%). Ethylene-propylene-diene monomer (EPDM) with an ethylene weight content of 70%, an unsaturated ratio DB/100c, and 8 and 22% propylene, was supplied by Hutchinson Technical Managing Engineering Department. The nitrile-butadiene rubber (NBR) was a Bayer product, Perbunan, which is a butadiene-acrylonitrile copolymer with 32% acrylonitrile content. The polychloroprene rubber (CR) was Bayprenllo (Bayer) with 37%  $\text{Cl}_2$  content. The poly(vinyl chloride) (PVC) was a BDH suspension polymer; it has a  $K$  value of 68. The peroxide used was 1,3-bis(isopropyl butyl)benzene (peroxide) on calcium carbonate (Perkadox 14/40, molecular weight 338) supplied by Hutchinson Technical Managing Engineering Department. The specifications of the different types of rubber are given in Table I.<sup>12</sup>

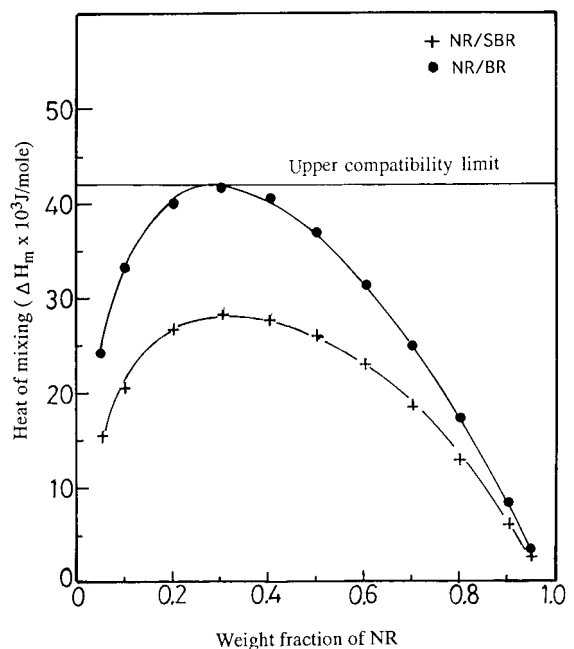


**Figure 2** The relationship between the permittivity ( $\epsilon'$ ) and the rubber content.

## Techniques

### Blend Preparation

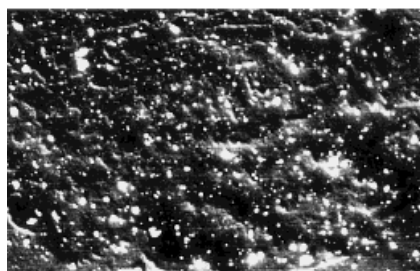
The blending of the components was carried out in a Brabender plasticorder at rotor speed of 30 rpm. The temperature of mixing was 150°C. The mixing was continued for 5 min, and then the peroxide was added to the mix on a laboratory two-roll mill (470-mm diameter, 300-mm working distance). The speed of the slow roll was 24 rpm and the gear ratio was 1 : 1.4. The compounded blends were left overnight before vulcanization.



**Figure 3** The relationship between the heat of mixing and the weight fraction of NR.



NR/SBR (X=500)



NR/BR (X=500)

**Figure 4** Scanning electron micrographs of NR/SBR and NR/BR rubber blends (original magnification  $\times 500$ ).

### Vulcanization

The vulcanization was carried out in a heated platten press under a pressure of about 40 kg/cm<sup>2</sup>

and a temperature of  $152 \pm 1^\circ\text{C}$  for their optimum cure time ( $t_{c90}$ ).

### Testing of Blend Mixes and Vulcanizate

The rheometric characteristics of the minimum torque ( $M_L$ ), maximum torque ( $M_H$ ), optimum cure time ( $t_{c90}$ ), scorch time ( $t_{s2}$ ), and cure rate index (CRI) were determined using a Monsanto 100 oscillating disk rheometer.

### Physicomechanical Measurements

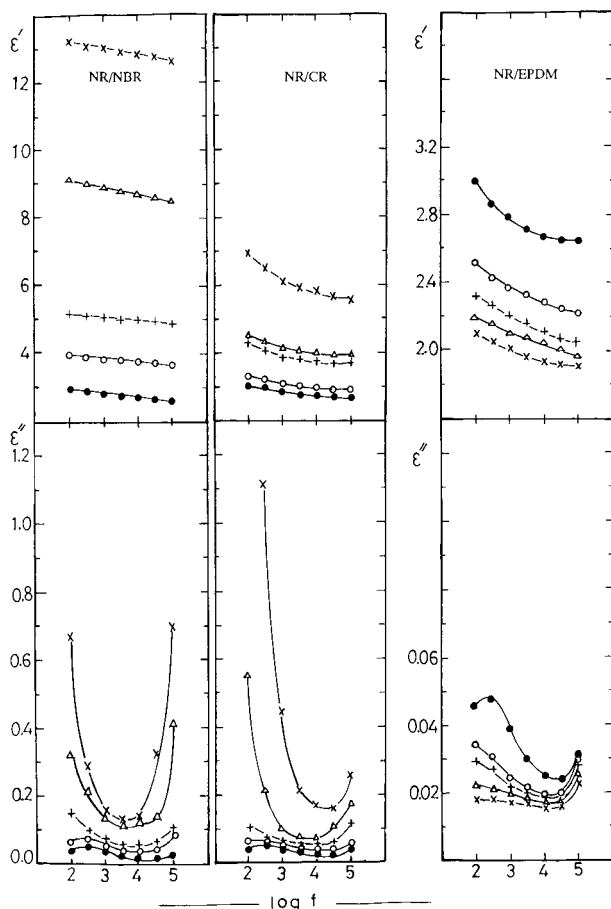
Tensile strength, modulus, and elongation at break were measured on seven dumbbell specimens for each sample according to ASTM D412-66T using a computerized (Zwick 1425) test instrument at a crosshead speed of 500 mm/min. Swelling<sup>13</sup> was carried out in toluene at room temperature ( $\sim 27^\circ\text{C}$ ) for 24 h and the percentage soluble fraction<sup>14</sup> was determined.

### Scanning Electron Microscopy Testing

Phase morphologies were studied using a Jeol JSM-T20 scanning microscope. For scanning electron observation the surface of the polymer was mounted on a standard specimen stub. A thin coating ( $\sim 10^{-6}$  m) of gold was deposited onto the

**Table III** Rheometric Characteristics and Physicomechanical Properties of NR/NBR, NR/CR, and NR/EPDM Blends

	Sample												
	A	C <sub>1</sub>	C <sub>2</sub>	C <sub>3</sub>	C <sub>4</sub>	D <sub>1</sub>	D <sub>2</sub>	D <sub>3</sub>	D <sub>4</sub>	E <sub>1</sub>	E <sub>2</sub>	E <sub>3</sub>	E <sub>4</sub>
Formulation													
NR	100	75	50	25	—	75	50	25	—	75	50	25	—
NBR	—	25	50	75	100	—	—	—	—	—	—	—	—
CR	—	—	—	—	—	25	50	75	100	—	—	—	—
EPDM	—	—	—	—	—	—	—	—	—	25	50	75	100
Peroxide	4	4	4	4	4	4	4	4	4	4	4	4	4
Rheometric characteristics at $152 \pm 1^\circ\text{C}$													
$M_L$ (dN m)	6.5	1.75	1.75	3	4.25	4.5	4	4.73	6.75	9.75	5.25	4.5	1.5
$M_H$ (dN m)	40	44	53	56	59	48	40	37	23	65	50	43	65
$t_{s2}$ (min)	2.5	4	4	4.5	3.4	2.5	2.25	2.5	3.5	3	2.5	3	2.75
$t_{c90}$ (min)	40.5	42.5	41.5	42	45	37.5	23	28.5	35	36	38.5	37	34.5
CRI (min <sup>-1</sup> )	2.63	2.59	2.66	2.66	2.40	2.85	4.82	3.84	3.17	3.03	2.77	2.94	3.14
Physicomechanical properties													
$M$ at 100% $E$													
(MPa)	0.36	0.34	0.52	0.56	0.59	0.51	0.39	0.35	0.21	0.59	0.51	0.58	0.68
TS (MPa)	1.01	1.71	1.38	3.28	1.21	1.73	4.75	1.89	1.2	3.34	3.49	3.6	3.14
Elongation (%)	225	300	238	488	229	300	450	340	350	750	688	625	700
Equilibrium swelling (%)													
	310	304.7	280	280.5	250	325	382	334	379	451	538	678	451
Soluble fraction (%)													
	3.53	2.06	2.72	3.34	2.39	3.76	5	6.48	6.5	4.8	5.1	7.2	4.2



**Figure 5** The permittivity ( $\epsilon'$ ) and dielectric loss ( $\epsilon''$ ) versus the frequency ( $\log f$ ) for NR/NBR, NR/CR, and NR/EPDM rubber blends with (●) 100, (○) 75, (+) 50, (Δ) 25, and (×) 0 parts of NR in the blend.

polymer surface and attached to the stub prior to examination in the microscope to enhance the conductivity and secondary electron emission characteristic of the overgrowth.

#### Dielectric Measurements

The permittivity ( $\epsilon'$ ) and dielectric loss ( $\epsilon''$ ) for the investigated samples were measured at frequencies ranging from 100 Hz to 100 kHz. An LCR meter (AG-4311B, Ando Electric Ltd) with an NFM/5T test cell was used. The capacitance  $C$  and the loss tangent ( $\tan \delta$ ) were obtained directly from the bridge from which  $\epsilon'$  and  $\epsilon''$  were calculated. The samples were in the form of disks of 58-mm diameter and 3-mm thickness. Calibration of the apparatus was carried out using standard samples (Trolitul, glass, and air). The accuracy for  $\epsilon'$  was  $\pm 1\%$  and for  $\epsilon''$  was  $\pm 2\%$ . The

measurements were carried out at 20°C using an ultrathermostat.

#### Conductivity Measurements

The electrical conductivity ( $\sigma$ ) of the investigated samples was measured by the application of Ohm's law using the NFM/5T test cell. A power supply unit (GM 45161/01, Philipps) was used. The potential difference  $V$  between the plates holding the sample and the current  $I$  flowing through it was measured by a multimeter (URI 1050, Rohde and Schwarz). The electrical conductivity was calculated using the equation

$$\sigma = \frac{dI}{AV} \text{ mho m}^{-1}$$

where  $d$  is the thickness of the sample (m) and  $A$  is its surface area ( $\text{m}^2$ ).

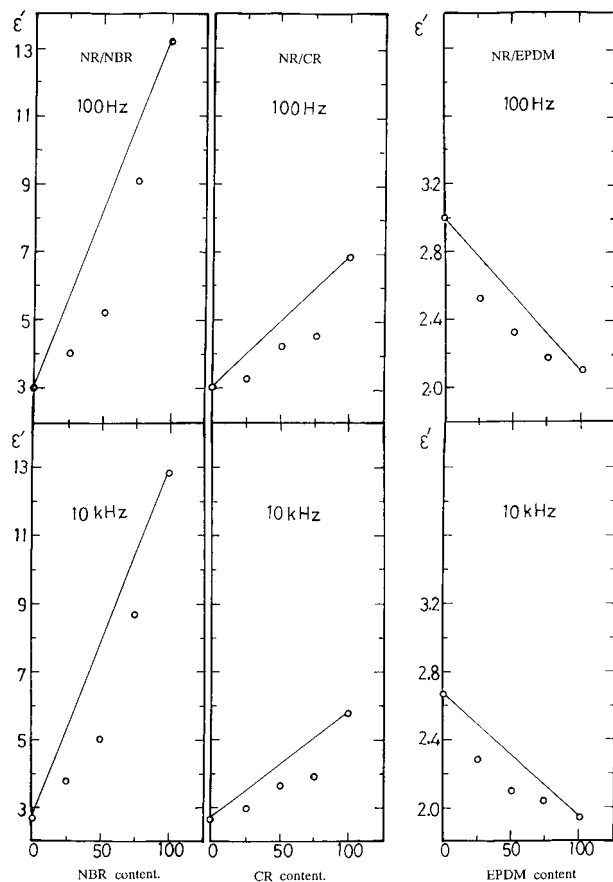
#### RESULTS AND DISCUSSION

One of the most important and known methods for modification of polymeric materials is the blending of polymers. In this study NR was blended with different synthetic rubbers in different ratios (100/0, 75/25, 50/50, 25/75, and 0/100) and the electrical and physicochemical properties of these blends were studied.

#### NR/SBR and NR/BR Blends

The NR was blended with SBR or BR as nonpolar types. The rheometric characteristics were determined at  $152 \pm 1^\circ\text{C}$  and the obtained mixes were vulcanized at their optimum cure times. The physicochemical properties were also determined and the obtained data are reported in Table II.

The values of the  $\epsilon'$  and  $\epsilon''$  for the different concentrations of the previous blends were studied over the frequency range from 100 Hz to 100 kHz at room temperature ( $\sim 27^\circ\text{C}$ ). The results obtained for  $\epsilon'$  and  $\epsilon''$  versus the frequency for these blends are shown in Figure 1. It is evident from this figure that  $\epsilon'$  decreases with increasing the applied frequency and shows an anomalous dispersion. From the absorption curves relating  $\epsilon''$  and  $\log f$  it is found that there is a peak whose maximum lies at around 300 Hz as shown in Figure 1. This peak is independent of the concentration of the polymer blends, leading to either dc

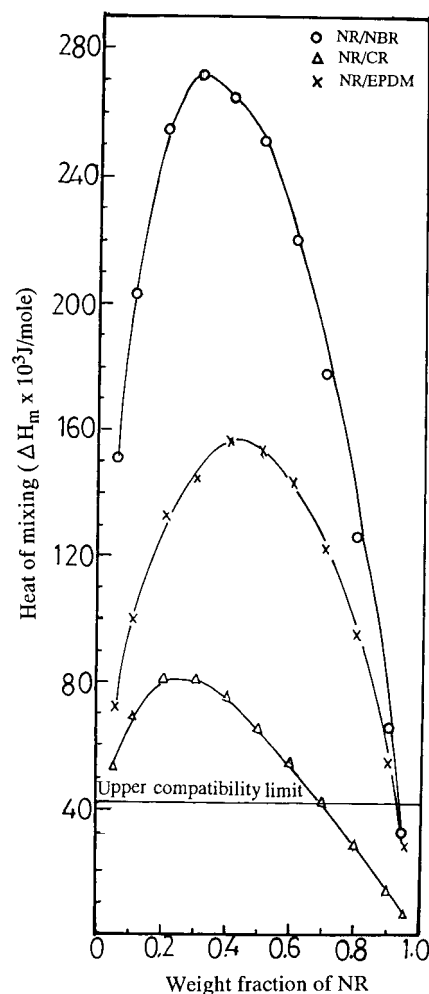


**Figure 6** The relationship between the permittivity ( $\epsilon'$ ) and the rubber content.

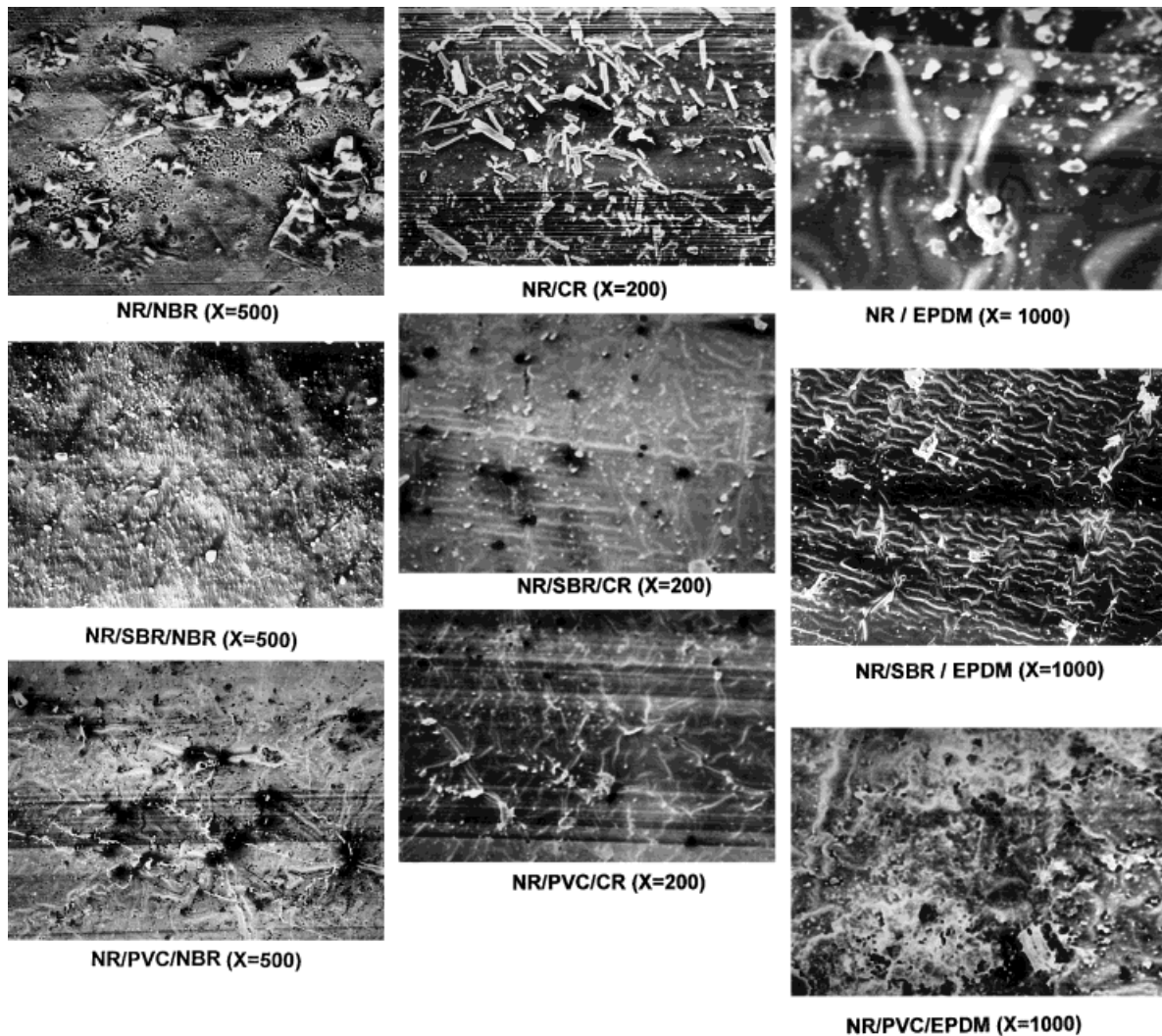
conductivity or the Maxwell–Wagner effect. No direct current was seen to flow through the samples, indicating that there was no dc conductivity. Moreover, it was ascertained that this effect was not due to a bad contact between the sample and the condenser plates because the measurements were repeated with aluminum foil stuck to the two faces of the samples and no change in the results was noticed. Hence, it is certain that the absorption region detected in that frequency range is attributed mainly to the Maxwell–Wagner effect. The origin of this effect is an ac current which is in phase with the applied potential. This current results from the difference in conductivities and permittivities of rubber and peroxide.<sup>15</sup> In addition, the absorption spectrum still shows a tail of a high frequency absorption region with a maximum higher than 100 kHz, which could be attributed to the mobility of the main chain and its related motions.<sup>16</sup>

In order to test the compatibility between NR and SBR or BR, the  $\epsilon'$  was plotted graphically in

Figure 2 versus the content of rubber in the blend at different frequencies (100 Hz, 1 kHz, 10 kHz, and 100 kHz). From this figure it is clear that the values of  $\epsilon'$  for NR/SBR and NR/BR blends coincide with the line connecting NR and SBR or BR. This linearity may indicate that these blends are compatible through the whole investigated range of concentrations. This presumption could be justified through the calculated heat of mixing<sup>10</sup> of the previous blends over the entire range of weight percent composition given in Figure 3. It was reported<sup>17</sup> that the heat of mixing can be taken as an approximate measure of the free energy of mixing and may be used as an indicator of possible compatibility. Singh and Singh<sup>11</sup> calculated the heat of mixing of some polymer blends using the Schneier equation<sup>10</sup> and reported that the heat of mixing with values below a limiting



**Figure 7** The relationship between the heat of mixing and the weight fraction of NR.



**Figure 8** Scanning electron micrographs of NR/NBR, NR/CR, and NR/EPDM rubber blends before and after the addition of SBR and PVC compatibilizers (original magnifications  $\times 500$ ,  $\times 200$ , and  $\times 1000$ ).

value of  $4.185 \times 10^{-2}$  J/mol indicates compatible blends while the incompatible blends have mostly higher values. Therefore, these blends are expected to be thermodynamically compatible because the calculated heats of mixing at all compositions shown in Figure 3 are below the limiting value of compatibility. These thermodynamic calculations may indicate a significant degree of compatibility, which was confirmed from the morphology using the electron microscope for the 50/50 NR/SBR and NR/BR blends as shown in Figure 4. The figure shows that these blends seem to be compatible because dark and bright regions representing both polymers are distributed homogeneously and none of the regions have phase separation.

#### NR/NBR, NR/CR, and NR/EPDM Blends

In this study NR was blended with NBR, or CR as polar rubbers and EPDM as a nonpolar one. The rheological characteristics were determined at  $152 \pm 1^\circ\text{C}$  and the obtained mixes were vulcanized at their optimum cure time. The physico-mechanical properties were also determined and the obtained data are given in Table III. For the rheometric characteristics of NR/EPDM blend it was observed that the  $M_L$  and  $M_H$  of EPDM are higher than those of NR. This may be attributed to the high viscoelastic plasticity behavior of EPDM.

The permittivity and dielectric loss for the three types of rubber blends with different ratios

**Table IV Rheometric Characteristics and Physicomechanical Properties of NR/SBR/NBR, NR/SBR/CR, and NR/SBR/EPDM Blends**

	Sample												
	F	F <sub>1</sub>	F <sub>2</sub>	F <sub>3</sub>	F <sub>4</sub>	N <sub>1</sub>	N <sub>2</sub>	N <sub>3</sub>	N <sub>4</sub>	M <sub>1</sub>	M <sub>2</sub>	M <sub>3</sub>	M <sub>4</sub>
Formulation													
NR	100	75	50	25	—	75	50	25	—	75	50	25	—
NBR	—	25	50	75	100	—	—	—	—	—	—	—	—
CR	—	—	—	—	—	25	50	75	100	—	—	—	—
EPDM	—	—	—	—	—	—	—	—	—	25	50	75	100
SBR	10	10	10	10	10	10	10	10	10	10	10	10	10
Peroxide	4	4	4	4	4	4	4	4	4	4	4	4	4
Rheometric characteristics at 152 ± 1°C													
$M_L$ (dN m)	4.5	5.75	6	4	5	5.5	5	6	7	5	6.5	9.0	9.5
$M_H$ (dN m)	39	57	62	62.5	71	57.5	48	47.5	46	59	65	79	81
$t_{s2}$ (min)	3	3	3	3.25	3.5	2.75	2.5	2	1.75	3.125	2.75	2.5	2.375
$t_{c90}$ (min)	39	38	38.5	38.5	38.5	37.5	36.5	36	38	38	37.5	37	36
CRI (min <sup>-1</sup> )	2.77	2.857	2.82	2.83	2.85	2.87	2.94	2.94	3.007	3.29	2.87	2.89	2.9
Physicomechanical properties													
$M$ at 100% $E$													
(MPa)	0.75	0.357	0.355	0.351	0.21	0.52	0.60	0.62	0.57	0.87	0.916	1.39	1.27
TS (MPa)	1.1	4.2	3.19	3.1	1.74	2.1	3.53	1.41	1.4	3.31	3.85	4.71	4.4
Elongation (%)	270	600	625	600	500	400	583	225	225	380	400	325	350
Equilibrium													
swelling (%)	264.6	260.6	275	198	185	310	328	265	290	245	255	202	330
Soluble													
fraction (%)	4.49	5.45	5.96	4.32	4.2	4.66	6.07	3.1	5.1	4.25	4.12	3.07	4.7

were measured over the frequency range from 100 Hz to 100 kHz at room temperature ( $\sim 27^\circ\text{C}$ ). The obtained data are illustrated graphically in Figure 5. From this figure it is noticed that the  $\epsilon'$  for the NR/NBR blend is much higher than NR/CR and NR/EPDM blends, which may be attributed to its higher polarity. The absorption curves relating  $\epsilon''$  and  $\log f$  (Fig. 5) show more than one relaxation process. The loss region could be resolved into two separate regions, a low absorption region with a maximum at a frequency lower than 100 Hz due to the Maxwell–Wagner effect and a tail of a higher frequency region with a maximum higher than 100 kHz due to the mobility of the main chain and its related motions.

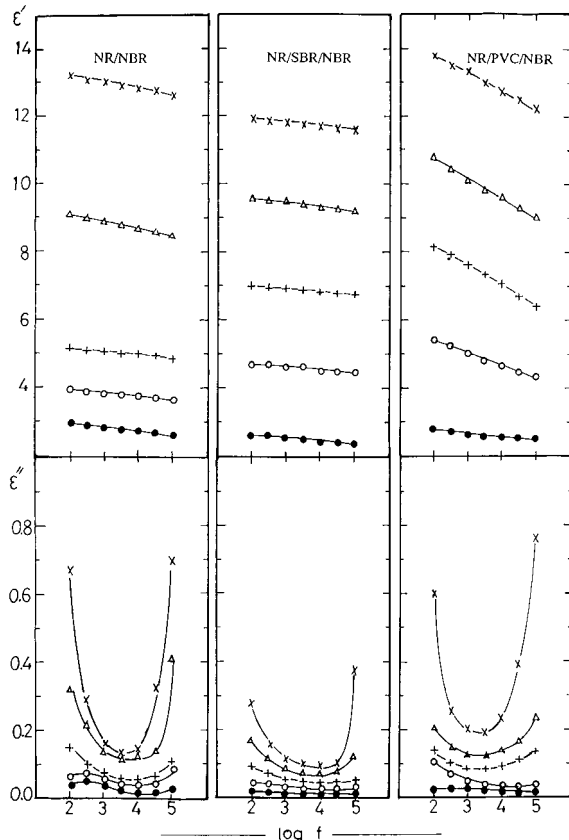
To check the compatibility between NR with NBR, CR, or EPDM, the  $\epsilon'$  is plotted graphically in Figure 6 versus the content of rubber in the blend at frequencies of 100 Hz and 10 kHz. This figure shows a deviation of the  $\epsilon'$  of the blends from the line connecting the values of both NR and NBR, CR, or EPDM. This nonlinearity may be attributed to the incompatibility of these blends. This result is supported by the calculation of the heat of mixing given in Figure 7. This figure gives the relation between the theoretical calculated heat of mixing and the composition of the

NR percentage. The calculated values of the heat of mixing for these blends are found to lie above the limiting value of compatibility,<sup>11</sup> especially for NR/NBR blend, which may indicate that these blends are thermodynamically incompatible blends. This incompatibility may be due to the difference in polarity between the two homopolymers and was confirmed using a scanning electron microscope on the 50/50 NR/NBR, NR/CR, and NR/EPDM blends as shown in Figure 8. From this figure it is clear that some etching is apparent for the NR/EPDM blend whereas the morphology of NR/NBR and NR/CR blends is shown as platy-shaped domains dispersed in the matrix with the distinguished interfacial boundaries between the domain and matrix. This observation indicates that these blends are heterogeneous (incompatible).

### Compatibilization

The most important parameter determining the quality of a blend is the degree of compatibility of the components to form one homogeneous phase with intermediate properties. From the previous results it is evident that these systems are incompatible blends. In other words, there is phase

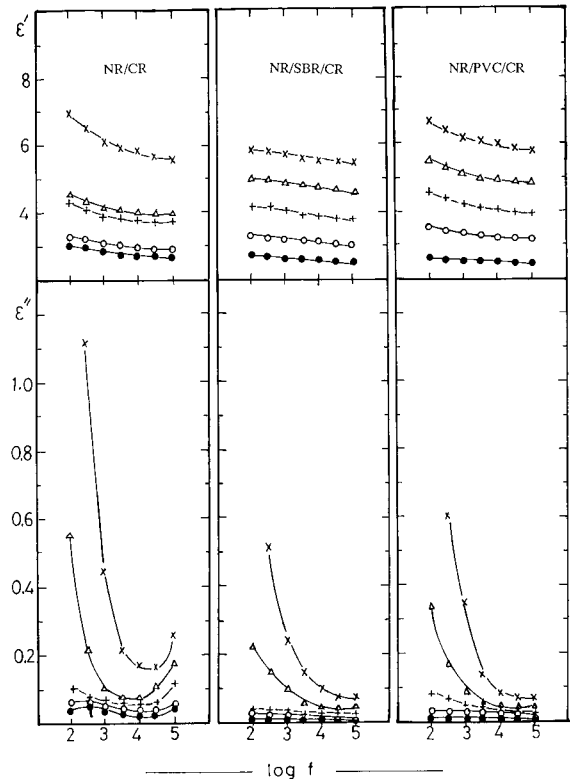




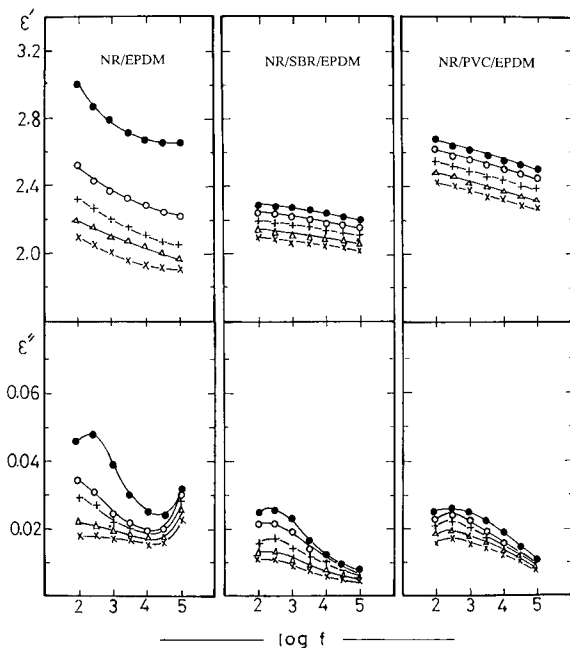
**Figure 9** The permittivity ( $\epsilon'$ ) and dielectric loss ( $\epsilon''$ ) versus the frequency ( $\log f$ ) for NR/NBR, NR/SBR/NBR, and NR/PVC/NBR rubber blends with (●) 100, (○) 75, (+) 50, (Δ) 25, and (×) 0 parts of NR in the blend.

separation between the two rubbers in these systems. In order to overcome the problem of phase separation, some trials were made to improve the compatibility of NR with either NBR, CR, or EPDM. The addition of a third component compatible with the two other ones and preferentially located at the interface between the two phases could be an easy way to improve the compatibility between two intrinsically incompatible rubbers. For this purpose, SBR and poly(vinyl chloride) (PVC)<sup>16</sup> (10 phr) were used as compatibilizing agents to enhance the compatibility of the above binary rubber blends.

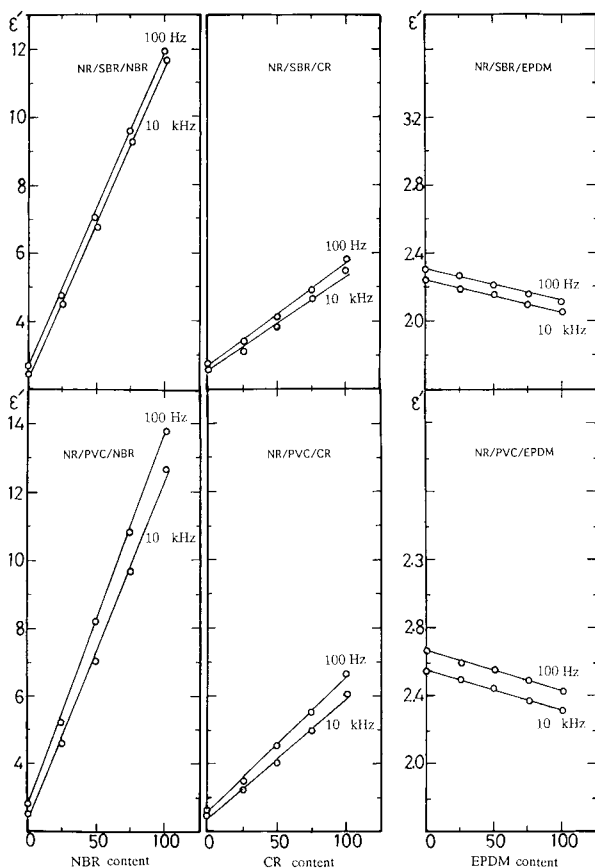
The formulations of the previous blends with SBR, the rheometric characteristics at  $152 \pm 1^\circ\text{C}$ , and their physicomechanical properties are given in Table IV. From the obtained data it is clear that incorporation of SBR in the blends produces a marked improvement in the tensile strength and modulus of these blends. These results may indicate that SBR enhances the adhesion between



**Figure 10** The permittivity ( $\epsilon'$ ) and dielectric loss ( $\epsilon''$ ) versus the frequency ( $\log f$ ) for NR/CR, NR/SBR/CR, and NR/PVC/CR rubber blends with (●) 100, (○) 75, (+) 50, (Δ) 25, and (×) 0 parts of NR in the blend.



**Figure 11** The permittivity ( $\epsilon'$ ) and dielectric loss ( $\epsilon''$ ) versus the frequency ( $\log f$ ) for NR/EPDM, NR/SBR/EPDM, and NR/PVC/EPDM rubber blends with (●) 100, (○) 75, (+) 50, (Δ) 25, and (×) 0 parts of NR in the blend.



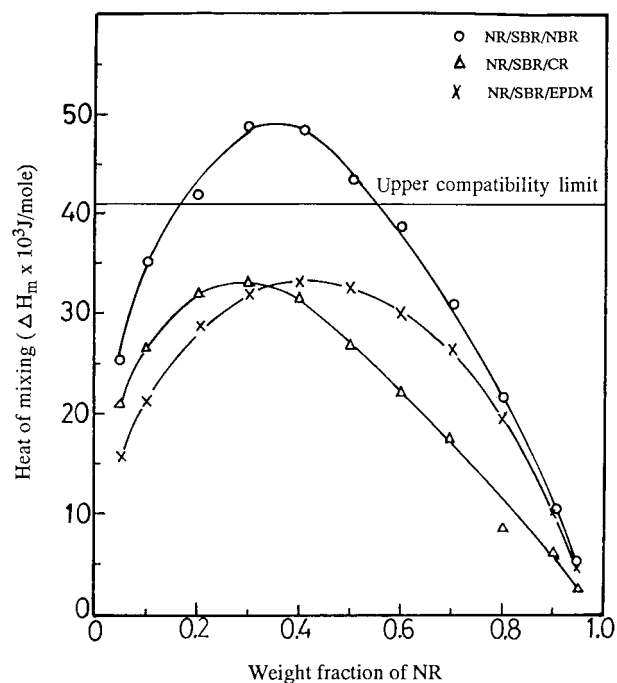
**Figure 12** The relationship between the permittivity ( $\epsilon'$ ) and the rubber content.

the two phases and minimizes the phase separation.<sup>18</sup>

The results obtained for  $\epsilon'$  and  $\epsilon''$  versus the frequency for these incompatible blends before and after the addition of the compatibilizer are shown in Figures 9–11. From these figures it is clear that the shape of the absorption curves was changed after the addition of the compatibilizer. On the other hand, Figure 12 represents the variation of  $\epsilon'$  with the content of the parent rubber in NR/SBR/NBR, NR/SBR/CR, and NR/SBR/EPDM blends at frequencies of 100 Hz and 10 kHz. It is clear that the  $\epsilon'$  value of the blends containing SBR lies on the straight line connecting the two individuals for the two types of rubber. This means that the addition of SBR as a compatibilizer may improve the compatibility of these blends to some extent. This conclusion is in good agreement with the results of the heat of mixing of NR/SBR/NBR, NR/SBR/CR, and NR/SBR/EPDM blends (Fig. 13), which show that the heat of mixing is lower than the upper limit of compatibility for NR/SBR/CR and NR/SBR/EPDM blends

whereas it is slightly higher than the limit value of compatibility for the NR/SBR/NBR blend, indicating that the incorporation of SBR as a compatibilizer partially improves the compatibility of the incompatible NR/NBR blend. This conclusion is also confirmed by the morphology of the investigated blends given in Figure 8, which indicates that the addition of SBR to NR/NBR and NR/CR blends loosens the fibrils and hence the surface becomes more uniform and the granular structure is more prominent. On the other hand, the addition of SBR to NR/EPDM leads to the disappearance of the holes and the reduction of the particle size of the dispersed phase. These results clearly indicate that the addition of SBR improves the interaction between the phases and thereby slows down the phase separation process.

Table V represents the formulations of the incompatible blends with PVC and the rheometric characteristics at  $152 \pm 1^\circ\text{C}$ , as well as their physicochemical properties. It is evident that the addition of PVC improves the mechanical properties, especially for the NR/PVC/NBR blend. This may be attributed to the compatibility of the PVC/NBR resulting from the existence of strong dipole interactions due to the polar acrylonitrile group. This compatibility leads to intermolecular diffusion across the interface.<sup>19</sup>



**Figure 13** The relationship between the heat of mixing and the weight fraction of NR after the addition of SBR.

**Table V Rheometric Characteristics and Physicomechanical Properties of NR/PVC/NBR, NR/PVC/CR, and NR/PVC/EPDM Blends**

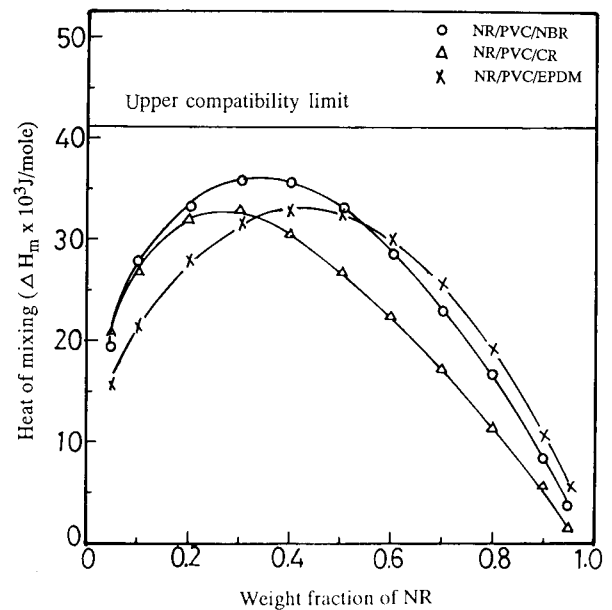
	Sample												
	F'	F' <sub>1</sub>	F' <sub>2</sub>	F' <sub>3</sub>	F' <sub>4</sub>	N' <sub>1</sub>	N' <sub>2</sub>	N' <sub>3</sub>	N' <sub>4</sub>	M' <sub>1</sub>	M' <sub>2</sub>	M' <sub>3</sub>	M' <sub>4</sub>
Formulation													
NR	100	75	50	25	—	75	50	25	—	75	50	25	—
NBR	—	25	50	75	100	—	—	—	—	—	—	—	—
CR	—	—	—	—	—	25	50	75	100	—	—	—	—
EPDM	—	—	—	—	—	—	—	—	—	25	50	75	100
PVC	10	10	10	10	10	10	10	10	10	10	10	10	10
Peroxide	4	4	4	4	4	4	4	4	4	4	4	4	4
Rheometric characteristics at 152°C ± 1													
M <sub>L</sub> (dN m)	5.5	2.5	3	5	7	3.5	4	5.5	11	5	10	13	21
M <sub>H</sub> (dN m)	54	54	25.5	49	59	56	53	42.5	38	58	64.75	74.5	73
t <sub>s2</sub> (min)	2.5	3.25	2.5	5	4	3.25	2.25	2	2	2.5	2.875	2.75	3.25
t <sub>c90</sub> (min)	33.5	34	25	31	33	29.5	20.5	18	30	36	34.5	35	34.5
CRI (min <sup>-1</sup> )	3.23	3.25	4.44	3.85	3.44	3.81	5.48	6.45	3.22	2.99	3.16	3.10	3.2
Physicomechanical properties													
M at 100% E													
(MPa)	0.573	0.655	0.38	0.68	0.89	0.35	0.4	0.702	0.81	0.45	0.68	0.84	0.79
TS (MPa)	1.72	2.83	8.69	4.62	5.35	2.71	4.46	3.08	4.17	3.6	5.24	3.86	4.79
Elongation (%)	150	225	595	250	350	300	475	375	350	225	250	250	300
Equilibrium swelling (%)	248	232	347.8	176	163	306	272	261	286	247	284	226	227
Soluble fraction (%)	3.79	4.51	4.2	2.83	1.39	4.95	3.53	1.03	6.06	3.56	3.39	3.27	3.13

The variation of  $\varepsilon'$  with the content of rubber in NR/NBR, NR/CR, and NR/EPDM blends after the addition of PVC at frequencies of 100 Hz and 10 kHz is also represented in Figure 12. A linear relationship was obtained that may indicate a greater degree of compatibility by adding PVC as a compatibilizing agent. This conclusion was confirmed by the values of the heat of mixing (Fig. 14), which lie below the upper limit of compatibility for the three blends. This is further confirmed by the micrograph shown in Figure 8, which indicates that the addition of PVC to those blends improves the phase morphology where the domains appear to be more uniformly dispersed. This can be attributed to the location of the PVC at the interfaces between the major phases and around the microphases.

## CONCLUSION

From the dielectric measurements, the calculation of the heat of mixing and the scanning electron microscopy, it was concluded that NR/SBR and NR/BR blends are compatible whereas NR/EPDM, NR/NBR, and NR/CR blends are incom-

patible. This leads to the conclusion that the dielectric method is well suited to study the degree of compatibility, as well as the heat of mixing and



**Figure 14** The relationship between the heat of mixing and the weight fraction of NR after the addition of PVC.

the scanning electron microscopy testing. On the other hand, the degree of compatibility of incompatible blends was improved by the addition of SBR and PVC to these blends.

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