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Investigations on the addition of styrene butadiene rubber in natural rubber and dichlorocarbene modified styrene butadiene rubber blends

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This paper focuses on the use of styrene butadiene rubber (SBR) as a viscosity modifier in novel blends of natural rubber (NR) and dichlorocarbene modified styrene butadiene rubber (DCSBR). The processing characteristics, vulcanisation kinetics, stress-strain behaviour, mechanical properties and low temperature transition of the blends have been examined in order to analyse the influence of SBR in the blends. The change in cross-link density values from stress strain behaviour and equilibrium swelling data has been correlated with the technological properties of the blends. The excellent mechanical properties and the increased cross-link density in blends in the presence of 5–10 phr of styrene butadiene rubber reveals the viscosity modifying action of SBR in NR/DCSBR blends. The variation in viscosities of these blends with the addition of SBR is reflected in the DSC thermograms. The resulting blends show very high resistance to thermal ageing as compared to those without SBR. © *2002 Kluwer Academic Publishers*

1. Introduction

Blending of two or more polymers for property improvement and economic advantage has gained considerable importance. A large number of polymer blends has been proposed for commercial purpose [1], all of them do not have the required mechanical properties due mainly to either their incompatibility or viscosity mismatch or other related problems. This arises because of the absence of specific interaction between the phases in such blends, which can be significantly improved by adding suitable ingredients during processing [2–6]. In the case of compatibilisers the segments can be chemically identical with those in the respective phases [7–9] or miscible with or adhere to one of the phases [10, 11]. According to Paul [12] this type of surface activity should reduce interfacial energy between the phases, permit a finer dispersion during mixing, provide a measure of stability against gross segregation [13] and result in improved interfacial adhesion [14]. A compatibilised blend hence gives a more homogeneous crosslink density during vulcanisation, which results in enhanced technological properties. A third component can successfully acts as a compatibiliser only if it contains segments in it which are identical to the components forming the blends. In other cases the third component can act either an inert additive or a viscosity modifier. The main steps after the development of novel elastomers and blends is the analysis on the processing aspects. This is because a very good understanding of various processing aspects is most needed for the conversion of a material from less useful to a more useful shape (moulding). In order to have optimum rheological characteristics, the viscosity of a system must be controlled carefully by the addition of viscosity modifiers for example use of chloroprene rubber in paint industry. Rubber blends based on natural rubber (NR) are extensively used in several potential applications. NR is used as a component in tire tread because of its excellent mechanical properties. Natural Rubber differs from other elastomers mainly is showing strain crystallisation nature and can therefore offers good physical properties. But it suffers from poor weathering, ozone, oil and thermal resistance. These properties result from its high content of

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unsaturation. Dichlorocarbene modified styrene butadiene rubber (DCSBR) can provide excellent oil, heat, flame, ozone and good compression set resistance [15]. The objective of the present work is to investigate the processing, thermal and mechanical properties of NR/DCSBR blends containing SBR as third component and to analyse the effect of loading of SBR on 70/30, 50/50 and 30/70 composition of NR/DCSBR blends.

2. Experimental

2.1. Materials

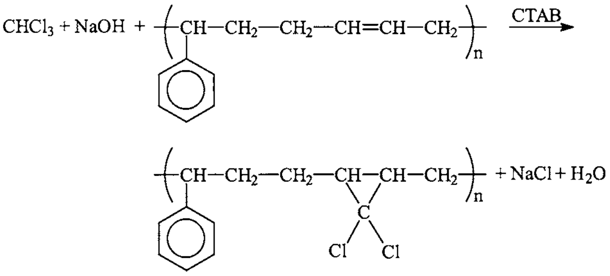
The basic materials used in this work are given below.

1. Natural rubber used for the study was ISNR-5 grade rubber, obtained from Pilot Crumb Rubber Factory, Rubber Board, Kottayam, Kerala, India.
2. DCSBR, dichlorocarbene modified styrene butadiene rubber was synthesized in the laboratory.
3. Other compounding ingredients such as zinc oxide, stearic acid, CBS (*N*-cyclohexyl-2-benzothiazyl sulphenamide), TMTD (tetra methyl thiuram disulfide), TDQ (2,2,4-trimethyl 1,2-dihydroquinoline), sulphur etc. were of reagent grade and obtained from local rubber chemical suppliers.

2.2. Methods

2.2.1. Preparation of DCSBR

Dichlorocarbene modified styrene butadiene rubber with 25% chlorine content was prepared by the alkaline hydrolysis of chloroform using cetyl trimethyl ammonium bromide (CTAB) as phase transfer catalyst as reported earlier [16]. The structural changes take place during chemical modification is shown in Scheme 1.



Scheme 1 Structural changes takes place during chemical modification of SBR.

2.2.2. Mixing

Master batches of NR and DCSBR were prepared separately and then blended on a laboratory size two roll-mixing mill having a friction ratio 1:1.4, as per ASTM D 15-627. SBR was added to the preblended NR/DCSBR (70/30, 50/50, 30/70) at a dosage of 5, 10 and 15 phr. The basic formulation used in the study is given in Table I.

2.2.3. Rheometry

The cure characteristics of the mixes were determined using a Monsanto Rheometer model R-100 at 150°C by measuring the optimum cure time and scorch time according to ASTM D 2705.

2.2.4. Preparation of test samples

The compounds were then compression moulded at 150°C using as electrically heated hydraulic press to their respective cure time (t_{90}). Dumbbell shaped tensile and angular tear specimens was punched out from the compression-moulded slabs along the mill grain direction.

2.2.5. Differential scanning calorimeter (DSC)

The glass transition behaviour of 50/50 NR/DCSBR blend was done on a Perkin-Elmer differential scanning calorimeter, operated at a heating rate of 15°C/min within the temperature range of -100 to 50°C.

2.2.6. Scanning electron microscopy (SEM)

Scanning electron microscopic studies of fractured samples were prepared by fracturing the ebonite treated samples in liquid nitrogen. The cut edges of the samples were taken in a JEOL scanning electron microscope (SEM).

2.2.7. Physico-mechanical testing of the samples

Stress-strain data were determined on a Zwick Universal Testing Machine (UTM), using c-type dumbbell specimen, according to ASTM D 412–80. The tear strength was determined as per ASTM D 624-81 using angular tear specimens. Both the tests were done at 28°C and at a crosshead speed of 500 mm/min.

TABLE I Basic formulation of NR/DCSBR blends

			70/30 NR/DCSBR (phr)			50/50 NR/DCSBR (phr)				30/70 NR/DCSBR (phr)		
Ingredients	NR	DCSBR	N7	N7A	N7B	N5	N5A	N5B	N5C	N3	N3B	N3C
NR	100	–	70	70	70	50	50	50	50	30	30	30
DCSBR	–	100	30	30	30	50	50	50	50	70	70	70
SBR	–	–	–	5	10	–	5	10	15	–	5	10
Zinc oxide	5	5	5	5	5	5	5	5	5	5	5	5
Stearic acid	2	2	2	2	2	2	2	2	2	2	2	2
TDQ	1	1	1	1	1	1	1	1	1	1	1	1
CBS	0.8	1.2	0.8	0.8	0.8	0.8	0.8	0.8	0.8	0.8	0.8	0.8
TMTD	–	0.8	0.3	0.3	0.3	0.4	0.4	0.4	0.4	0.5	0.5	0.5
Sulphur	2.5	2.2	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2	2.2	2.2

The compression set values were determined by constant strain method keeping 22 hours for 70°C, according to ASTM D 395 (1982). The rebound resilience percentage values were determined using a Dunlop Tripsometer, as per ASTM D 1504. The abrasion resistance of the samples was tested using a DIN abrader according to DIN no. 5351 test method.

2.2.8. Thermal ageing

The resistance to thermal ageing of the samples was analysed by keeping the samples in an air oven at 70°C for 96 hours, as per ASTM D-573 procedure. The retained values of percentage in tensile strength and elongation at break were calculated.

2.2.9. Cross-link density

The cross-link density of a rubber vulcanisate can be estimated from swelling measurement, using the Flory-Rehner equation [17–19] or by a mechanical method involving stress strain measurement [20].

The cross-link density can be determined from swelling data, the samples were allowed to swell in toluene and the equilibrium uptake is noted. The molecular weight between the crosslink M_c is calculated using the following equation.

$$\frac{1}{2M_c} = \left[\frac{-\rho_r V_s (V_r)^{1/3}}{\ln(1-V_r) + V_r + \chi^2 V_r^2}\right] \tag{1}$$

where M_c = molecular weight of polymer between two crosslinks, ρ_r = density of polymer, V_s = molar volume of solvent and V_r = volume fraction of polymer in swollen mass is calculated by the method of Ellis and Welding [21] is given by

$$V_r = \frac{(d - f_w)\rho_r^{-1}}{(d - f_w)\rho_r^{-1} + A_s \rho_s^{-1}} \tag{2}$$

where A_s be the amount of solvent absorbed, ρ_r and ρ_s are the density of rubber and solvent respectively, d is the deswollen weight of the sample and f_w be the fraction of insoluble components.

The interaction parameter χ which is given by Hildebrand [22, 23] equation as

$$\chi = \beta + \frac{V_s}{RT}(\delta_s - \delta_P)^2 \tag{3}$$

where β = lattice constant; V_s = molar volume; R = universal gas constant; T = absolute temperature; δ_s = solubility parameter of solvent; δ_P = solubility parameter of polymer.

From molecular weight between crosslinks M_c, the crosslink density ν was calculated using the following equation [24].

$$\nu = \frac{1}{2M_c} \tag{4}$$

On the basis of phenomenological theory of rubber elasticity and derived from the Mooney Rivlin equation [25], stress-strain measurement can be used for measuring the crosslink density of rubber. This can be obtained using Equations (5) and (6) below. From the plot of $\sigma_0/(\lambda - \lambda^{-2})$ and $1/\lambda$, the constants C_1 and C_2 can be determined, the intercept of the curve on the $\sigma_0/(\lambda - \lambda^{-2})$ axis corresponds to the C_1 value and its slope corresponds to the value of C_2.

$$F = 2A_0(\lambda - \lambda^{-2})(C_1 + C_2\lambda^{-1}) \tag{5}$$

$$\sigma_0 \big/ (\lambda - \lambda^{-2}) = 2C_1 + 2C_2/\lambda \tag{6}$$

Where F is the tensile extension force required for stretching a specimen, A_0 is the cross-sectional area of the unstretched specimen, σ_0 is identifiable with F/A_0 and C_1 and C_2 characteristic constant of the vulcanisate. C_1 is directly related to the physically effective crosslink density (ν_{phys}) by the equation,

$$C_1 = \rho RT \nu_{phys} \tag{7}$$

3. Results and discussion

3.1. Cure characteristics

The rheographs of the NR/DCSBR blends containing SBR in different blend compositions are given in Figs 1–4 and the curing process data are presented in Table II. The lowest value of maximum viscosity is observed for DCSBR while comparatively higher value for NR. This is due to the comparatively increased difficulty for its shearing in rheometer. This results from the tendency for NR to show high elastic recovery, presence of nerves during shearing. Among the blends without SBR, as the NR content increases the maximum torque increases, this is again due to the same behaviour of NR as explained earlier. But in the case of samples with SBR, such an increasing trend with NR content is observed only for system with 10 phr SBR. In the case of samples with 5 phr SBR, the highest torque value is observed for 50/50 composition. In this case, the interpenetration of the blend is maximum which increases the maximum viscosity value. Considering a particular

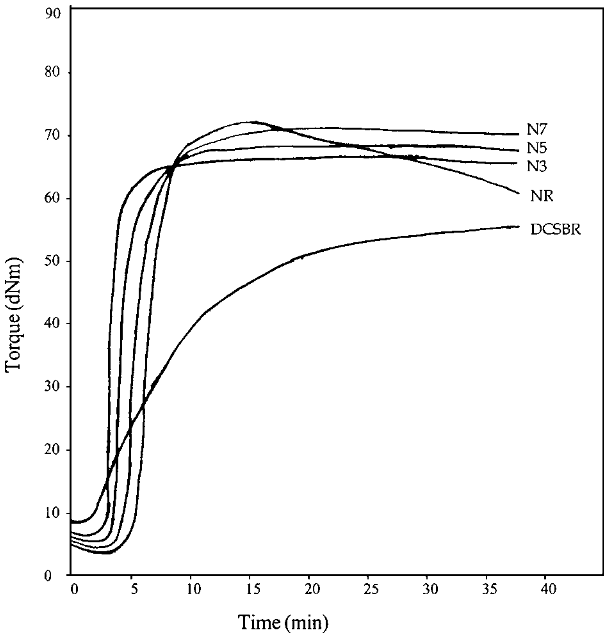


Figure 1 Rheographs of NR, DCSBR and NR/DCSBR blend with different composition (N7-70/30, N5-50/50 and N3-30/70) at 150°C.

TABLE II Cure characteristics of NR/DCSBR blends

Blend composition	Minimum torque (dNm)	Maximum torque (dNm)	Optimum cure time (t_{90}) (min)	Scorch time (t_2) (min)
NR	3	70	8	4
N7	4	68	8	3.5
N7A	4.5	70	7.5	3
N7B	5.5	72	7	2.5
N5	4.5	65	6.5	3
N5A	5	69	6	2.25
N5B	5.5	67	5.5	2
N5C	6	63	4.5	1.5
N3	7	63	5.5	2.5
N3A	8.5	60	4.5	2.25
N3B	10	58	3.5	2
DCSBR	8	51	14	2

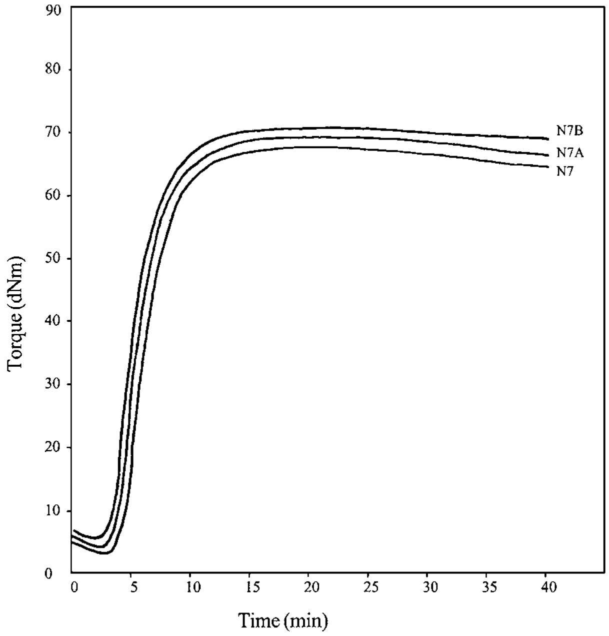


Figure 2 Rheographs of 70/30 NR/DCSBR blend containing (N7) 0, (N7A) 5 and (N7B) 10 phr SBR at 150°C.

blend composition, a regular decrease in maximum viscosities with increasing SBR content is observed only for 30/70 system, while for 50/50 and 70/30 system increasing trends are observed. It can be seen that the optimum cure time is maximum for DCSBR while with increase in concentration of NR in the blend, it initially decreases and then increases for 70/30 system. On the other hand scorch time shows a regular increase with increase in NR content. Normally optimum cure time and scorch time go hand in had in rheometric experiments. But here an irregular trend is observed. This means that the influence of NR on the curing process is mainly activated at intermediate and final stages of the curing process. It is already published in the literature that the pH of the system affects the curing properties to a considerable extent [26]. The presence of chlorine atom in DCSBR makes the system slightly more acidic than NR. This factor increases the optimum cure time. But this effect is activated only at later stages of vulcanisation, therefore a difference in the behaviour of optimum cure time and scorch time is observed. The increase in amount of SBR in the blend shows an initial increase and then a decrease in scorch time for all blend composition and the optimum cure time is found to be decreasing with increasing the concentration of SBR.

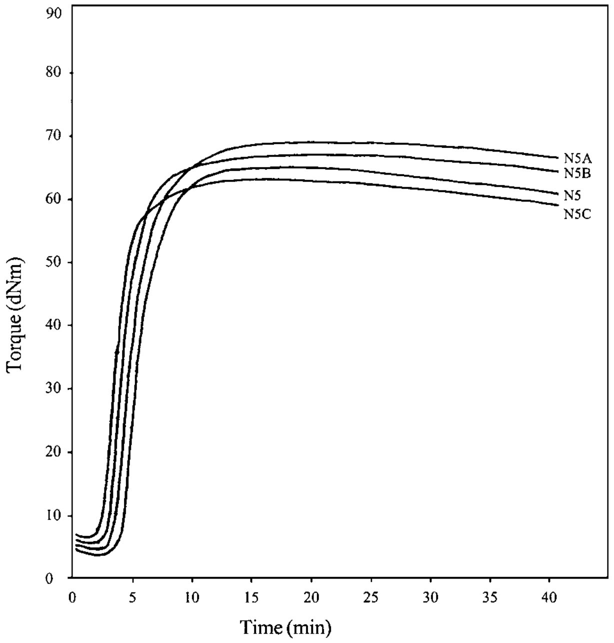


Figure 3 Rheographs of 50/50 NR/DCSBR blend containing (N5) 0, (N5A) 5, (N5B) 10 and (N5C) 15 phr SBR at 150°C.

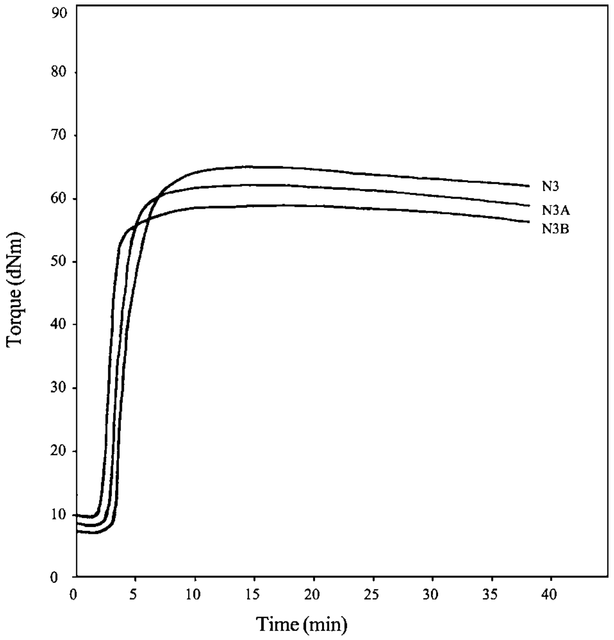


Figure 4 Rheographs of 30/70 NR/DCSBR blend containing (N3) 0, (N3A) 5 and (N3B) 10 phr SBR at 150°C.

3.2. DSC thermograms

DSC traces of 50/50 NR/DCSBR blend in the presence and absence of SBR is presented in Fig. 5. For 50/50 blends (N5), there appears two transitions, one at −68°C which correspond to the transition of NR and another at −55°C which relates to the DCSBR transition. But for 5 phr SBR containing 50/50 blend (N5A) the thermal transitions are shifted so that a single

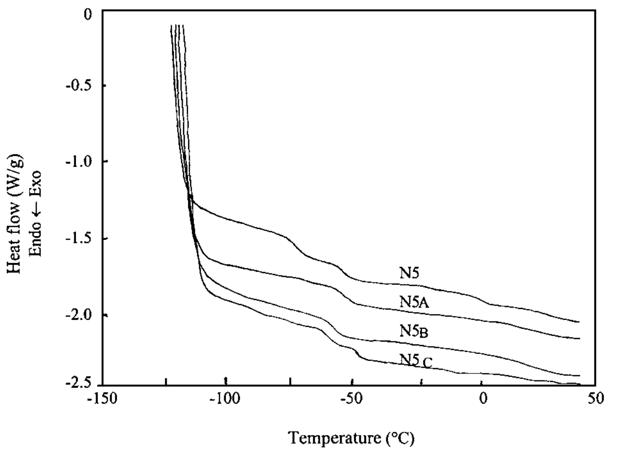


Figure 5 DSC thermograms of NR, DCSBR and 50/50 NR/DCSBR blend containing (N5) 0, (N5A) 5, (N5B) 10 and (N5C) 15 phr SBR.

transition at −60°C is observed. AS the amount of SBR in the blend increases, transition shift to −58°C (N5B). When the dosage of SBR become 15 phr (N5C) there is two thermal transition one at −62°C and other at −50°C. The forward shift in Tg of NR indicates the loss flexibility of NR with the addition of SBR while the backward shift in Tg of DCSBR remains the partial role of SBR as a plasticiser in DCSBR. So it can be understood that the addition of SBR causes a slightly opposing effect in NR and DCSBR, when the dosage of SBR becomes 15 phr. This indicates that SBR act as a third component revealing its identity. This can be made clear on examining the SEM photographs given below.

3.3. Scanning electron microscopy (SEM)

The SEM photographs of 50/50 NR/DCSBR blend having 5 to 15 phr loading of SBR is given in Fig. 6. Fig. 6a indicates the presence of NR as well as DCSBR which leads to two Tg's with the addition of 5 phr SBR, the enhancement in interpenetration of the blend component resulting in earlier stated shift in Tg. This is supported by the smooth surface of the sample (Fig. 6b). But when the concentration of SBR increases further, a tendency for SBR to agglomerates forming separate domains in the blend is clear (marked area of Fig. 6c).

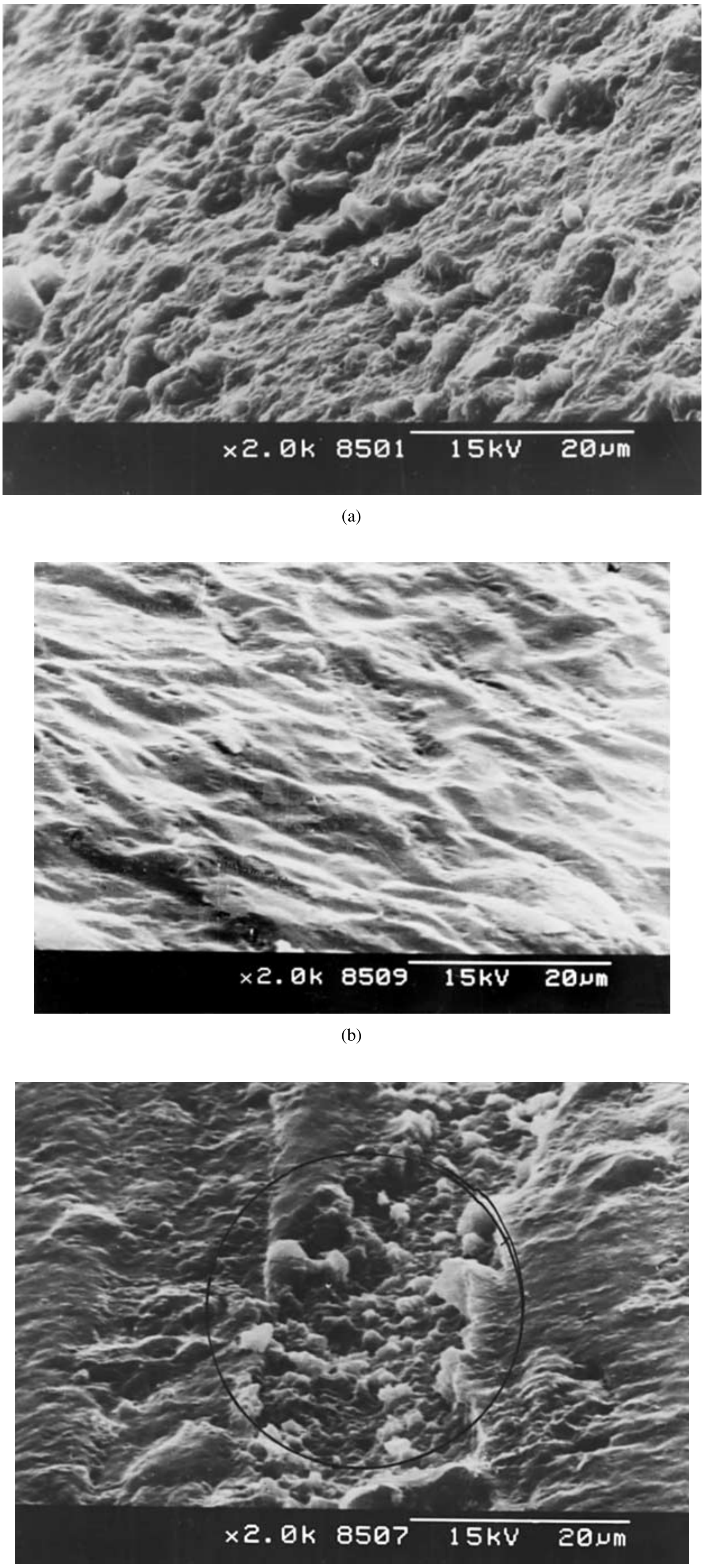
3.4. Effect of concentration of SBR on technological properties of NR/DCSBR blends Mechanical properties

The mechanical properties of the blends of NR/DCSBR are presented in Table III. An increase in tensile strength and elongation at break has been observed with increase in NR concentration in the blends. This behaviour suggests that each rubber in the blends has consumed balanced quantities of curatives to attain at a similar state of vulcanisation as a result of which a regular increase in physical properties with blend composition is observed. Addition of 5–10 parts of SBR greatly enhances the tensile properties of the blend when the concentration of DCSBR is lower or equal to NR. For 70/30 NR/DCSBR blend, higher values of tensile strength are obtained when concentration of SBR is 10 phr whereas in 50/50 composition, 5 phr loading of SBR gives maximum tensile strength. The improved tensile strength is due to the uniform distribution of the two rubber phases, SBR in the blend which facilitate efficient stress transfer to the two phases. At higher dosage of SBR, a decrease in tensile strength is obtained which is due to the higher amount of amorphous and weak SBR.

The superior tear strength is observed for 70/30 NR/DCSBR blends ratio, this observation points out that the tear strength is decided by stress factors such as the molecular packing, strength of the bonds in the material etc. in addition to the crosslink density of the sample. Blends with SBR containing samples shows higher tear strength as compared to pure blends (Table III). The superior tear strength is observed for 70/30 NR/DCSBR blend ratio, SBR creates efficient stress transfer leading to elongated DCSBR domains. The maximum tear strength is observed for 70/30 composition with 10 phr loading of SBR while that 5 phr loading is enough for 50/50 compositions. Compression set value is strongly dependent on the elastic recovery of the sample, it is seen from the Table III that the compression set of the blends increases with increase in concentration of NR. For SBR filed blends the percentage of compression set is lower than that of pure blends. As the loading of SBR increases the set decreases for 70/30 and 30/70 NR/DCSBR compositions. In 50/50 blends a lower set is noted for 5 phr SBR concentration. This is because the dispersed domains will respond differently in the stress-relaxation process (after compression period) and decreases the set values.

TABLE III Mechanical properties of NR/DCSBR blends

			70/30 NR/DCSBR (phr)			50/50 NR/DCSBR (phr)				30/70 NR/DCSBR (phr)		
Properties	NR	DCSBR	0	5	10	0	5	10	15	0	5	10
Tensile strength (MPa)	21.0	17.05	22.48	25.5	33.1	20.2	29.7	25.4	22.1	16.0	16.5	15.4
Modulus 300% (MPa)	2.89	3.9	3.09	3.63	3.71	3.88	3.77	3.99	3.58	3.51	1.63	3.86
Elongation at break (%)	854	524	849	891	792	764	711	678	634	551	498	531
Tear strength (KNm^{-1})	30.4	58.4	43.71	59.09	61.1	41	47.97	46.5	39.6	40.4	42.4	43.5
Resilience (%)	76.7	31	50.8	52	53	35.8	35	35	36	29.6	30	30.7
Compression set (%)	21.2	13.5	19.9	16.0	15.8	18	16	16.3	16.1	15.9	17.5	16.9
Heat build-up (°C)	10	11	12	12	11	13	14	13	13	15	18	14
Din abrasion loss (mm^3)	140.3	69.8	118.5	97.3	86.8	107.1	76.2	79.6	71.4	85.7	78	71.4



(a)

(b)

(c)

Figure 6 SEM of tensile fracture surfaces of 50/50 NR/DCSBR blend containing (a) 0, (b) 5 and (c) 15 phr SBR.

The abrasion resistance of NR/DCSBR blend is shown in Table III. It is seen from the table that blending of DCSBR increases the abrasion resistance, excellent abrasion resistance is observed for 30/70 NR/DCSBR blend composition. Compared to pure blends, SBR containing samples show a higher abrasion resistance. Similarly as the loading of SBR increases the abrasion resistance also increase.

The heat build-up of the samples which registers as the friction between the polymers which decrease with NR content increases (Table III), comparatively lower heat build-up value are noted for all blend composition at 10 phr loading of SBR than that of pure blends. Resilience values of NR/DCSBR blends increase with increase in concentration of NR. Lower resilience is observed for blends with SBR samples, but as the loading of SBR increases the resilience also increases (Table III).

The resistance of the rubber vulcanisate to thermal ageing is considered as an essential requirement for long service life of products. The percentage of decrease in tensile strength as a result of ageing at 70°C for 96 h is presented in Table IV. The ageing resistance decreases with increase in NR content. Comparatively higher ageing resistance is obtained for SBR containing samples. As the concentration of SBR, increases above 5 phr in 50/50 , a decrease in effect is observed.

TABLE IV Effect of air ageing on mechanical properties of NR/DCSBR blend at 70°C 96 h

Blend composition	Decrease in tensile strength (%)	Decrease in elongation at break (%)
NR	68.1	41.2
N7	59.9	39.5
N7A	53.1	38.3
N7B	51.3	36.0
N5	52.3	29.6
N5A	52.0	17.6
N5B	51.4	16.9
N5C	51.0	20.3
N3	39.6	24.5
N3A	40.3	14.9
N3B	40.0	12.6
DCSBR	12.3	14.4

From our investigation we suggest that the role of SBR in these blend be as a viscosity modifier. Owing to the absence of segments in SBR, which are identical to the blend components, it fails to act the role of a compatibiliser in these systems. It can be stated it is acting mearly as an interpenetrating agent that improves the viscosity of the system.

3.5. Calculation of crosslink density from swelling and stress-strain behaviour

The crosslink density values obtained from swelling data for NR/DCSBR blends are given in Table V. For pure blends the crosslink density is found to be decreasing with increase in concentration of NR. Higher crosslink density is observed for blends with SBR containing samples and as the dosage of SBR increases the crosslink density also increases. This is because of the restriction of swelling which causes an increase in V_r, which in turn increases the crosslink density.

The crosslink density values (ν_{phys}) calculated using the Equation 7 is given in Table V. It can be seen that the force and crosslink density is found to be maximum in 50/50 blend with 5 phr SBR and is in good agreement with the increase in rheometric toque. As the concentration of SBR increases above 5 phr, the $2C_2$ value is found to be decreasing (Table V). The higher $2C_2$ value for the 50/50 blend containing 5 phr SBR shows the presence of higher chain entanglement [27]. Since the elastomer matrix is composed of two components, a higher chain entanglement shows better molecular level mixing. The crosslink density increases in presence of SBR. Thus the observed tensile strength variation can be exactly correlated with the variation of ν_{phys} from Mooney-Rivlin equation and the crosslink density data from swelling studies. Moreover, the values of ν_{phys} are higher for blends with and without SBR samples compared to that of crosslink density from swelling studies. In swelling studies of these samples, crosslinks are flexible to penetrants, but as the loading of compatibiliser increases they restricts the swelling. The discrepancy between crosslink density determined by chemical analysis and by the application of stress-strain equation is partly due to the entanglements of interpenetrating network chains, which behave as crosslinks and partly

TABLE V Crosslink density parameters C_1, C_2, (ν_{phys}) from stress strain measurement, ν from swelling data and difference in rheometric torque of NR/DCSBR blends

Blend composition	Mooney-Rivlin equation × 10^{-3} (g molml^{-1})	Mh-Mn (dNm)	$2C_2$ (Nmm^{-2})	$2\,C_1$ (Nmm^{-2})	Flory-Rehner equation × 10^{-5} (g molml^{-1})
NR	3.04	67	4.15	0.896	3.1
N7	1.96	64	4.22	0.913	2.5
N7A	2.04	65	4.30	0.921	3.8
N7B	2.11	66.5	4.35	0.933	4.2
N5	1.75	60	4.09	0.888	3.9
N5A	1.99	64	4.39	0.910	4.8
N5B	1.84	61.5	4.31	0.902	4.3
N5C	1.69	57	4.01	0.870	4.0
N3	1.52	56	3.59	0.766	5.1
N3A	1.48	51.5	3.46	0.744	4.7
N3B	1.42	48	3.29	0.738	4.6
DCSBR	1.06	43	4.02	0.852	1.30

by the presence of elastically ineffective chain ends. This shows that there is sufficient molecular mixing of NR/DCSBR in presence of SBR.

4. Conclusions

SBR acts as an interpenetrating agents which improved the viscosity of natural rubber/dichlorocarbene modified styrene butadiene rubber. The presence of SBR in the blend is accompanied by an enhancement in technological properties, which depends on the composition of blend constituents and also on the concentration of SBR. The viscosity modifying action is more efficient in 70/30 and 50/50 blend composition. Results from differential scanning calorimetry showed that an appreciable extent of interpenetration was achieved in NR/DCSBR blends. The presence of chain entanglement as revealed from stress-strain isotherm is due to the better molecular level mixing and leads to improve mechanical properties.

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