

Improved Mechanical Properties of NR/EPDM Blends by Controlling the Migration of Curative and Filler via Reactive Processing Technique

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Received 16 March 2008; accepted 1 September 2008

DOI 10.1002/app.29193

Published online 4 November 2008 in Wiley InterScience (www.interscience.wiley.com).

ABSTRACT: Simple blending of natural rubber/ethylene-propylene-diene rubber (NR/EPDM) generally results in inferior mechanical properties because of curative migration and their differences for filler affinity. In this work, the 70/30 and 50/50 NR/EPDM blends prepared by reactive processing techniques were investigated and compared with the simple, nonreactive blends. The reactive blend compounds were prepared by preheating EPDM, containing all curatives to a predetermined time related to their scorch time prior to blending with NR. For the 70/30 gum blends, four types of accelerators were studied: 2,2-mercaptobenzothiazole (MBT), 2,2-dithiobis(benzothiazole) (MBTS), *N*-cyclohexyl-2-benzothiazolesulfenamide (CBS), and *N*-*tert*-butyl-2-benzothiazolesulfenamide (TBBS). When compared with the simple blends, the reactive blends cured with CBS and MBTS showed a clearly improved tensile strength whereas the increase of tensile strength in the blends cured with TBBS and MBT

was marginal. However, a dramatic improvement of ultimate tensile properties in the reactive 50/50 NR/EPDM blends cured with TBBS was observed when compared with the simple blend. For the *N*-550-filled blends at the blend ratios of 70/30 and 50/50, the reactive-filled blends prepared under the optimized preheating times demonstrated superior tensile strength and elongation at break over the simple blends. The improved crosslink and/or filler distribution between the two rubber phases in the reactive blends accounts for such improvement in their mechanical properties. This is shown in the scanning electron micrographs of the tensile fractured surfaces of the reactive blends, which indicate a more homogeneous blend. © 2008 Wiley Periodicals, Inc. *J Appl Polym Sci* 111: 2035–2043, 2009

Key words: blends; NR; EPDM; mechanical properties; reactive processing

INTRODUCTION

Blending of natural rubber (NR) and ethylene-propylene-diene rubber (EPDM) is done to combine the good mechanical and dynamic properties of NR and the heat and ozone resistance of EPDM for a broader range of applications. It has been demonstrated that inclusion of a sufficient amount of EPDM into NR or other unsaturated commodity rubbers such as BR and SBR improved their heat and ozone resistance.^{1–3} The finely dispersed phase of EPDM in highly unsaturated rubbers, provides an effective internal antiozonant equivalent to or even superior to conventional antiozonants such as *N*-(1,3-dimethylbutyl)-*N'*-phenyl-*p*-phenylenediamine (6PPD).^{1,4–6} However, it is

well recognized that blending of highly unsaturated NR and highly saturated EPDM generally leads to inferior mechanical properties of the blend vulcanizates caused by uneven distribution of crosslink or reinforcing filler or both.^{7–9} The differences in unsaturation levels lead to cure incompatibility especially in sulfur vulcanization systems. Moreover, highly polar accelerators preferentially partition in high unsaturated NR phase and promote a maldistribution of accelerator concentration and, as a consequence, the NR phase contains more crosslinks than the EPDM in the NR/EPDM blends.¹⁰ Utilization of different types of accelerators has been demonstrated to have an influence on mechanical properties of the simple NR/EPDM blends.¹¹ Various approaches have been adopted to overcome the cure incompatibility in blends of EPDM with diene rubbers as well as to get better filler distribution in such blends to optimize the blend properties. Those approaches included use of blend compatibilizers,^{2,12} modified EPDM,^{13–15} two-stage vulcanization,^{16,17} and a reactive blending technique.^{5,18} The latter technique has been proven to work very well with NR/BR/EPDM

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Contract grant sponsor: Thailand Research Fund (TRF) and Higher Education Commission; contract grant number: MRG4980043.

TABLE I
Compound Formulations of Various Accelerator Types

Ingredient	Part by weight			
	A	B	C	D
STR 20	70	70	70	70
Keltan 512	30	30	30	30
ZnO	4	4	4	4
Stearic acid	2	2	2	2
MBT ^a	0.67	–	–	–
MBTS ^a	–	1.33	–	–
CBS ^a	–	–	1.06	–
TBBS ^a	–	–	–	0.95
Sulfur	2.5	2.5	2.5	2.5

^a Accelerator contents equal to 4 mmol.

blends aimed to use for tire sidewall applications.⁵ In this work, the reactive processing technique was used to prepare the blends cured with sulfur vulcanization systems. The four different types of commonly used primary accelerators investigated in 70/30 NR/EPDM gum blends to determine the effectiveness of the technique were 2,2-mercaptobenzothiazole (MBT), 2,2-dithiobis- (benzothiazole) (MBTS), *N*-cyclohexyl-2-benzothiazolesulfenamide (CBS), and *N*-*tert*-butyl-2-benzothiazolesulfenamide (TBBS). The TBBS, which produced the optimum tensile properties in the gum blends, was later used for further investigation in the carbon black-filled blends at the same blend ratio, as well as in both gum and filled blends at the blend ratio of 50/50. The cure characteristics, mechanical properties, and surface topography of tensile fractured surfaces of all the reactive blends were assessed in comparison with those of the simple blends.

EXPERIMENTAL

Materials

NR (STR20) manufactured by Chana Latex Co., Ltd. (Songkhla, Thailand) and EPDM (Keltan 512) manufactured by DSM Elastomers B.V.(The Netherlands) were used for blend preparation. The EPDM rubber contains 4.3 wt % of ethylidene norbonene as a third monomer and 55 wt % of ethylene. Four different types of accelerators were used: MBT, MBTS, CBS, and TBBS. All were manufactured by Flexsys (Antwerpen, Belgium). The other compounding ingredients used were zinc oxide (Global Chemical, Thailand), stearic acid (Imperial Chemical, Thailand), and sulfur (Siam Chemical, Thailand). All elastomers and compounding ingredients were used as received.

Preparation of blend compounds

Initial Mooney viscosities, ML1 + 4 (125°C) of EPDM and NR tested according to ASTM D 1646

using Mooney Viscometer (SPRI, England) were 50 and 88, respectively. Prior to blending, NR was masticated on a two-roll mill to reduce its viscosity to be similar to that of EPDM. All the blend compounds were prepared by using a 6" × 14" two-roll mill with a friction ratio of 1 : 1.21. The simple and reactive blends of NR/EPDM were prepared with the same ingredients for comparison purposes.

Preparation of simple gum blend compounds

The simple 70/30 NR/EPDM blends with various accelerator types were prepared using the formulations given in Table I, and the simple 50/50 NR/EPDM blend cured with TBBS was compounded using the formulation shown in Table II. The pre-masticated NR was mixed with EPDM, then stearic acid, ZnO, accelerator, and sulfur were sequentially added, respectively. Total mixing time was 12 min. The blend compound was finally made into a 2–3 mm thick sheet by a two-roll mill.

Preparation of reactive gum blend compounds

The procedures used for preparing the reactive NR/EPDM blend compounds in this work were adapted from the reactive processing technique reported for NR/BR/EPDM blends elsewhere.¹⁸ The reactive 70/30 NR/EPDM blends using the four accelerator types and the reactive 50/50 NR/EPDM blend cured with TBBS were prepared using the formulations given in Tables I and II, respectively. The entire amounts of stearic acid, ZnO, accelerator, and sulfur were first sequentially incorporated into the EPDM only using a two-roll mill operated at room temperature. The EPDM compounds were tested for their scorch times at 150°C using an oscillating disk rheometer (ODR 2000, Monsanto) according to ASTM D 2084. Different preheating times for the EPDM were then assigned using scorch time as a reference point, as demonstrated in Figure 1. Preheating times assigned were in a range of few minutes prior to scorch time and would not exceed the scorch time to

TABLE II
Compound Formulations Using TBBS as an Accelerator

Ingredient	Part by weight		
	E	F	G
STR 20	50	50	70
Keltan 512	50	50	30
ZnO	4	4	4
Stearic acid	2	2	2
TBBS	0.95	0.95	0.95
N-550	–	50	50
Naphthenic oil	–	10	10
Sulfur	2.5	2.5	2.5

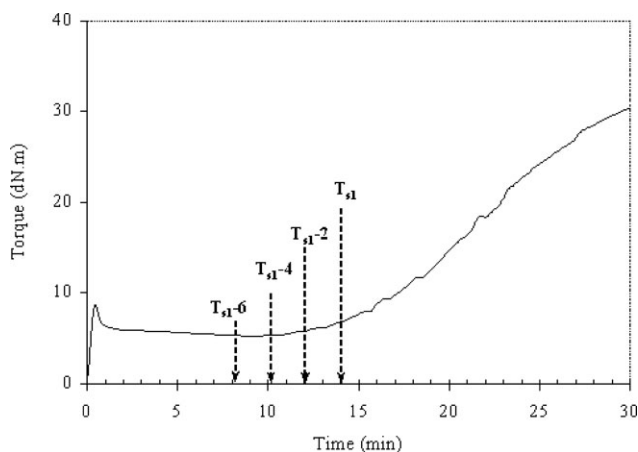


Figure 1 Example of preheating times assigned in reference to the scorch time of EPDM compound containing TBBS accelerator at 150°C.

keep the preheated EPDM processable. The EPDM compound was then preheated in a compression molding machine at 150°C for a predetermined time. The resulting preheated EPDM sheet was immediately removed from the mold, left to cool down to room temperature in an open atmosphere, and later blended with premasticated NR to a final required blend ratio.

Preparation of filled compounds

Two masterbatches of NR and EPDM with carbon black and oil were prepared. Each masterbatch consisted of rubber 100 parts, carbon black N-550 50 parts, and naphthenic oil 10 parts. For the simple-filled blends, the NR and EPDM masterbatches were mixed to the assigned final NR/EPDM blend ratios, i.e., 70/30 and 50/50 prior to adding stearic acid, ZnO, TBBS, and sulfur. For the reactive-filled blends, the EPDM masterbatch containing N-550 and oil was initially mixed with the entire amounts of stearic acid, ZnO, TBBS, and sulfur. The EPDM compound was made into a 2-mm-thick sheet, tested for its scorch time at 150°C, preheated to a time assigned related to the scorch time using a compression molding, and finally cross-blended with the masterbatch of NR to make up a final blend compound having all ingredients as shown in Table II.

Determination of curing characteristics and vulcanization

All blend compounds were tested for their curing characteristics, i.e., scorch time (T_{s1}), optimum cure time (T_{C90}), minimum torque (M_L), and maximum torque (M_H), using ODR according to ASTM D 2084 at 150°C. The compounds were subsequently press vulcanized to their optimum cure times, and the

resulting vulcanizates were left at room temperature for at least 16 h prior to being tested for their mechanical properties.

Determination of blend vulcanizate properties

Tensile properties

The blend vulcanizates were tested for their tensile properties, i.e., modulus at 100% elongation, tensile strength, and elongation at break according to ASTM D 412 at a crosshead speed of 500 mm/min, using Hounsfield Tensile Tester (H10KS, Hounsfield Test Equipment, England).

Crosslink density

Crosslink density of the vulcanizates was determined by equilibrium swelling method through Flory-Rehner equation.¹⁹ The samples were immersed in toluene at room temperature until swelling equilibrium occurred, i.e., until the swollen samples' weight remained constant. Prior to weighing, the samples were removed from the bottle and blotted with filter paper to remove excess toluene from the surface.

Morphological studies by SEM and TEM

Scanning electron microscopy (SEM) studies were carried out on the gold-coated tensile fractured surfaces of the blend vulcanizates by JSM 5410, JEOL. The magnification for all samples was at $\times 3500$. For the transmission electron microscopy (TEM) studies, 60 nm thin sections of the blend vulcanizates were cryogenically cut using an RMC model MTX ultramicrotome at -120°C . The nonstained sections were characterized using TEM (JEM 2010, JEOL) at a magnification of $\times 6000$.

RESULTS AND DISCUSSION

Curing characteristics and tensile properties of simple and reactive 70/30 NR/EPDM gum blends

The EPDM compounds with the four different types of accelerators, CBS, TBBS, MBT, and MBTS, displayed different scorch times at 150°C, and hence, preheating times for each compound were assigned differently. Four different preheating times were assigned for each EPDM compound. For example, the EPDM compound cured with the TBBS had a scorch time of 14 min, so the preheating times assigned were 12 ($T_{s1}-2$), 10 ($T_{s1}-4$), 8 ($T_{s1}-6$), and 6 ($T_{s1}-8$) min. The EPDM compound cured with the MBT had a scorch time of 2.5 min, therefore the preheating times assigned were 2 ($T_{s1}-0.5$), 1.5 ($T_{s1}-1$), 1 ($T_{s1}-1.5$), and 0.5 ($T_{s1}-2$) min. Different preheating

TABLE III
Cure Characteristics at 150°C of the 70/30 NR/EPDM Gum Blend Compounds

Accelerator type	Blend type	Preheating time ^a (min)	M_L (dN m)	$M_H - M_L$ (dN m)	Scorch time, T_{s1} (min)	Cure time, T_{c90} (min)
CBS	Simple	–	2.69	19.65	6.36	11.00
	Reactive	T_{s1-1}	2.66	20.28	3.90	8.70
TBBS	Simple	–	2.12	20.73	7.40	13.50
	Reactive	T_{s1-2}	2.88	20.22	2.81	8.87
MBT	Simple	–	2.81	14.76	1.79	11.00
	Reactive	$T_{s1-1.5}$	4.11	14.76	1.44	9.80
MBTS	Simple	–	2.26	15.53	4.32	10.50
	Reactive	T_{s1-3}	3.40	18.59	1.66	7.45

^a Preheating times that gave an optimum tensile strength of the blend vulcanizates.

periods were evaluated to find a preheating time which gave optimum tensile strength. Curing characteristics of the final blend compounds prepared with simple and reactive techniques are compared in Table III. For the reactive blends, the curing characteristics of only one representative blend compound are demonstrated here. The scorch times at 150°C of the EPDM cured with TBBS, CBS, MBT, and MBTS, which were used as reference points were 14, 12, 2.5, and 18 min, respectively.

The reactive blends clearly exhibited shorter scorch times and cure times in comparison with those of the simple blends for all accelerator types as shown in Table III. This was the result of the preactivated EPDM component during the preheating step. It is generally accepted that torque difference ($M_H - M_L$) from the cure curve is proportional to the extent of crosslinking in the rubber vulcanizate.¹¹ From the results shown in Table III, the reactive blends tend to show torque differences higher than those of the simple blend counterparts. So, this implies a higher crosslink density in the reactive blends in relation to the simple blends.

Tensile properties of the reactive gum blend vulcanizates cured with different types of accelerators

were investigated in comparison with the properties of the simple blends. For each accelerator type, the reactive blends prepared by using the preheated EPDMs, which had been subjected to various preheating times prior to blending with NR, were tested. It was found that the tensile strength of the reactive blends varied with varying preheating times, as typically shown in Figure 2 for the blends cured with the MBTS. It can be clearly seen from Figure 2 that the optimum tensile strength of the MBTS cured reactive 70/30 NR/EPDM blends was obtained when the preheating time of T_{s1-3} min was used. We found that the optimum preheating times for the EPDM compounds cured with CBS, TBBS, MBT, and MBTS were T_{s1-1} , T_{s1-2} , $T_{s1-1.5}$, and T_{s1-3} min, respectively. The tensile strength and elongation at break of the reactive blends prepared by using the EPDM compounds preheated to their optimum preheating times are shown in Figures 3 and 4, and compared with those of the simple blends. It was found that the reactive blends cured with MBTS and CBS showed significant improvement in tensile strength, whereas there was only a marginal increase in tensile strength when MBT and TBBS were used. The increase of ultimate tensile stress in the reactive

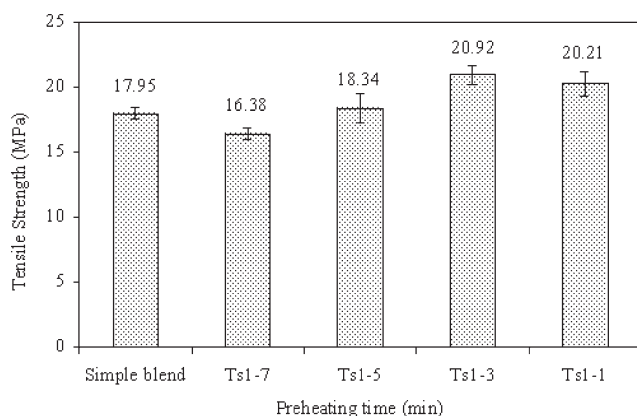


Figure 2 Tensile strength of the simple and reactive 70/30 NR/EPDM blends of varying EPDM preheating times cured with MBTS.

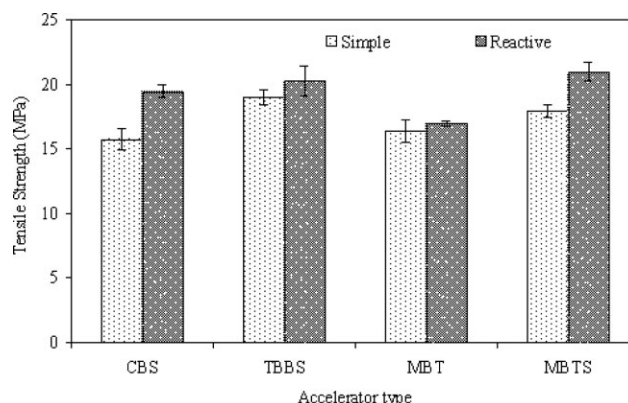


Figure 3 Tensile strength of the simple and optimum reactive 70/30 NR/EPDM blends.

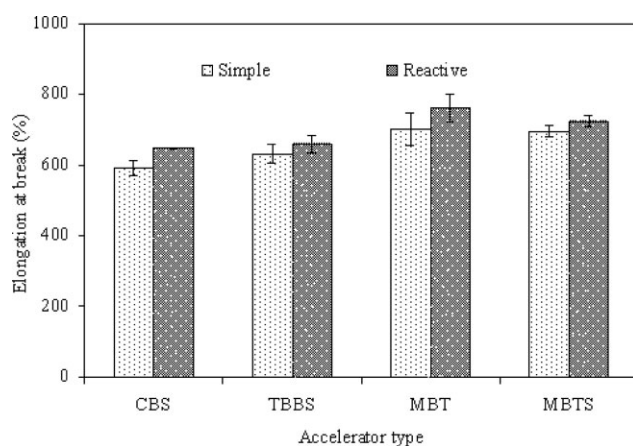


Figure 4 Elongation at break of the simple and optimum reactive 70/30 NR/EPDM blends.

blends does suggest an improved crosslink distribution in the blends prepared through reactive technique. This improvement in mechanical property could be attributed to a reduced curative migration, and hence, a better crosslink distribution in agreement with the work reported for the ternary blends of NR/BR/EPDM cured with the CBS accelerator.^{5,18} The preheating step of EPDM compounds resulted in accelerator fragments attaching to the EPDM rubber chains, and thereafter the curative migration from the EPDM phase toward the higher unsaturated NR phase was restricted, resulting in a more homogeneous curative distribution between the two rubber phases.

It is clear that the reactive technique was effective for such blends cured with CBS and MBTS, but less pronounced in the blends cured with MBT and TBBS, as exhibited in Figure 3. The TBBS was reported to provide the NR/EPDM blends with rather good cure compatibility¹¹ in agreement with our results for the simple blend. It can be seen in Figure 3 that the simple blend cured with TBBS depicted higher tensile strength than those cured with MBTS, MBT, and CBS. In Figure 3, the CBS was found to give the lowest tensile strength for the simple blends, among the four types of accelerators studied. This is believed to be caused mainly by its high cure rate, low melting temperature (97°C), and high polarity, which resulted in a greater amount of accelerator migration and hence maldistribution of accelerator concentration between the NR and EPDM phases.

The crosslink density of the simple and reactive blends with an optimum tensile strength determined by equilibrium swelling method in toluene is shown in Table IV. This reveals that the reactive blends contain more crosslinks than the simple blends, except the blends with MBT where both types of blends showed similar crosslink density. The increased

crosslink density could be attributed to more crosslinks in the EPDM phase that lead to an improvement in crosslink distribution between each blend component. Characterization of crosslink distribution in rubber blends is not a simple task and has no absolute method. The similar solubility parameters of NR and EPDM (16.5 and 16.1 MPa^{1/2}, respectively) introduce even more difficulties. Some of the techniques such as swollen state NMR and differential swelling methods applied for polar/nonpolar rubber blend²⁰ require the difference in solubility of each blend component in each selected solvent, so that the techniques are not applicable to the NR/EPDM blends. Sophisticated solid-state NMR is probably the most suitable technique, however, the technique is complicated and requires expertise. We expect to implement such a technique, to get more insight about local crosslink distribution in the blends, and the outcome shall be reported in our future publications.

The TBBS was found to provide the best overall tensile properties in the 70/30 NR/EPDM blends prepared by both simple and reactive techniques, as indicated in Figure 3. The blends demonstrated good mechanical strength because the blends are composed of 70 parts of NR. To further evaluate the effectiveness of reactive processing technique on such blends, the blends with lower content of NR, i.e., higher content of EPDM, were also worth considering. The NR/EPDM blends at the blend ratio of 50/50 cured with TBBS were therefore investigated.

Curing characteristics and tensile properties of simple and reactive 50/50 NR/EPDM gum blends

The blend compounds were prepared following the formulation given in Table II, utilizing both simple and reactive blending techniques. The reactive blends were prepared by using the EPDM, which was preheated to various times predetermined in reference to its scorch time. The reactive blends showed shorter scorch and cure times than those of the simple blends, as shown in Table V. A reduction of scorch and cure times of the reactive blend was due to the utilization of the preactivated EPDM, as previously discussed for the case of the 70/30 NR/EPDM blends.

TABLE IV
Crosslink Density in the Simple and Optimum Reactive 70/30 NR/EPDM Gum Blends

Blend type	Crosslink density (mol/m ³)			
	CBS	TBBS	MBT	MBTS
Simple	173 ± 6	175 ± 4	123 ± 3	137 ± 2
Reactive	176 ± 5	181 ± 3	122 ± 2	146 ± 2

TABLE V
Cure Characteristics of the Simple and Reactive 50/50 NR/EPDM Gum and Filled Blends with TBBS Accelerator
(the EPDM Preheating Time = T_{s1} -1.5 min for the Reactive Blends)

Blend type	Cure property at 150°C				
	M_L (dN m)	M_H (dN m)	$M_H - M_L$ (dN m)	Scorch time (min)	Cure time (min)
Gum-Simple	3.57	22.91	19.34	7.60	16.31
Gum-Reactive	3.29	24.11	20.82	2.28	9.78
Filled-Simple	5.93	39.44	33.51	4.00	12.50
Filled-Reactive	5.44	36.19	30.75	1.75	10.50

Figure 5 clearly shows that the EPDM preheating time has an influence on the tensile strength of the 50/50 NR/EPDM reactive blends. This behavior was previously observed for the 70/30 NR/EPDM blends, as demonstrated in Figure 2. For the 50/50 NR/EPDM gum reactive blend, the optimum preheating time was found to be equal to T_{s1} -1.5 min. On comparing the tensile strength of the 70/30 and 50/50 NR/EPDM blends cured with the TBBS as shown in Figures 3 and 5, respectively, it is clear that increasing the EPDM contents from 30 to 50 parts in the blends resulted in a significant reduction in tensile strength of the blend vulcanizates. This is obviously due to a poor strength of the unfilled EPDM vulcanizate. However, as illustrated in Figures 5 and 6, it was found that the utilization of the reactive processing technique remarkably improved the tensile properties of the NR/EPDM blends.

When the two blend components have similar viscosities, the blending of NR with EPDM at the blend ratio of 50/50 could result in a cocontinuous blend morphology.²¹ At this 50/50 blend ratio, the curative migration from highly saturated EPDM to highly unsaturated NR could lead to very inferior mechanical properties, because the high strength NR would no longer act as a matrix. The results shown in Figure 6 revealed that both tensile strength and elongation at break of the simple 50/50 NR/EPDM blend are very much lower than those of the reactive blend pre-

pared under the optimum preheating condition. Such improvement is inevitably influenced by a better crosslink distribution in the reactive blends. The reactive blend also displayed more crosslinks in comparison with the simple blend, as indirectly indicated by the torque differences in Table V.

Tensile properties of N-550-filled simple and reactive NR/EPDM blends at the blend ratios of 70/30 and 50/50

The improvement of ultimate tensile properties by the reactive processing technique has so far been demonstrated in the nonfilled or gum blend vulcanizates. To evaluate the effectiveness of the technique, the carbon black-filled blends of NR/EPDM having the blends ratios of 70/30 and 50/50 were prepared according to the formulations given in Table II. The preheating time of the filled EPDM compound that gave the optimum ultimate tensile properties was found at T_{s1} -1.5 min. The tensile properties of the 70/30 and 50/50 NR/EPDM blends filled with 50 phr of N-550, prepared by simple and reactive techniques, are compared in Figures 7 and 8, respectively.

As demonstrated in Figures 7 and 8, the reactive blends at both blend ratios revealed a great improvement in ultimate tensile properties. The improved mechanical properties are attributed to a better

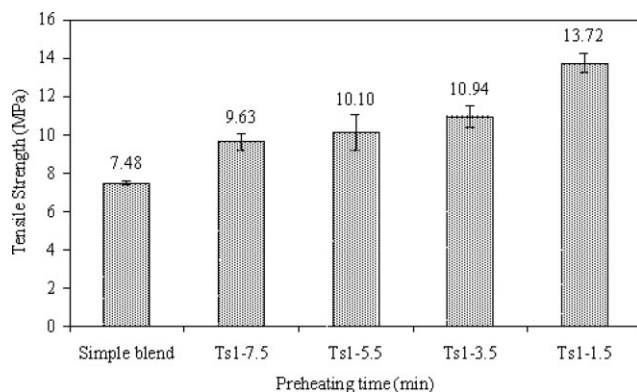


Figure 5 Tensile strength of the simple and reactive 50/50 NR/EPDM gum blends of varying EPDM preheating times cured with TBBS.

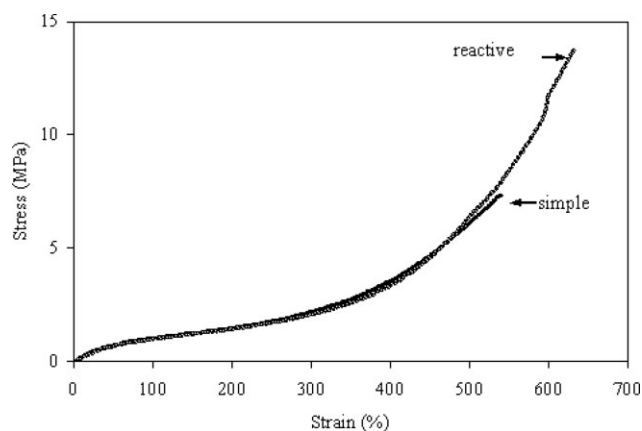


Figure 6 Stress-strain curves of the simple and reactive (T_{s1} -1.5) 50/50 NR/EPDM gum blends with TBBS accelerator.

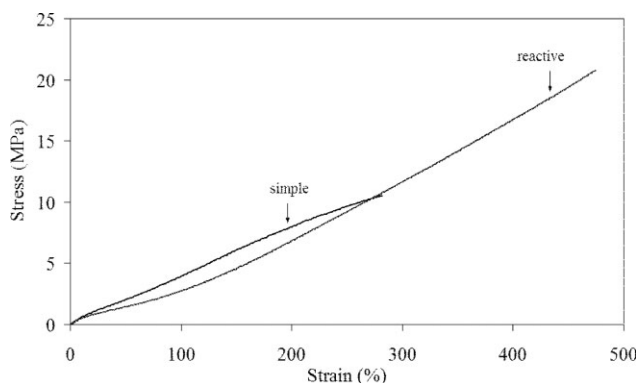


Figure 7 Stress–strain curves of the simple and reactive (T_{s1} -1.5) 70/30 NR/EPDM blends filled with 50 phr of *N*-550.

crosslink distribution, as discussed earlier for the results of gum blend vulcanizates. In addition, the EPDM-bound intermediate, and possibly a few crosslinks, formed after the preheating step would retard migration of carbon black toward the NR phase and result in a better balance of filler distributed between the two phases.

Morphological studies of the simple and reactive NR/EPDM blends

The improved ultimate tensile properties of the gum and *N*-550-filled NR/EPDM blends at the blend

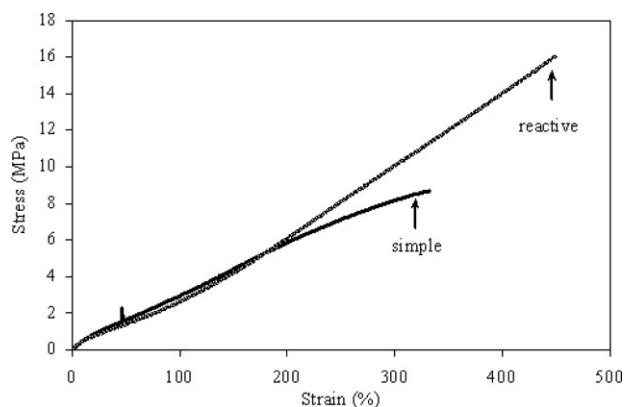


Figure 8 Stress–strain curves of the simple and reactive 50/50 (T_{s1} -1.5) NR/EPDM blends filled with 50 phr of *N*-550.

ratios of 70/30 and 50/50 prepared by reactive processing technique are in good agreement with the results observed in SEM studies. Figures 9 and 10 show SEM micrographs of the tensile-fractured surfaces of the NR/EPDM blends at the blend ratios of 70/30 and 50/50 containing no (gum) and 50 phr of carbon black *N*-550, respectively. The reactive blend specimens subjected to this characterization were those of the blends prepared under the optimum preheating times, which gave the best tensile strength. The fracture surfaces of the reactive gum

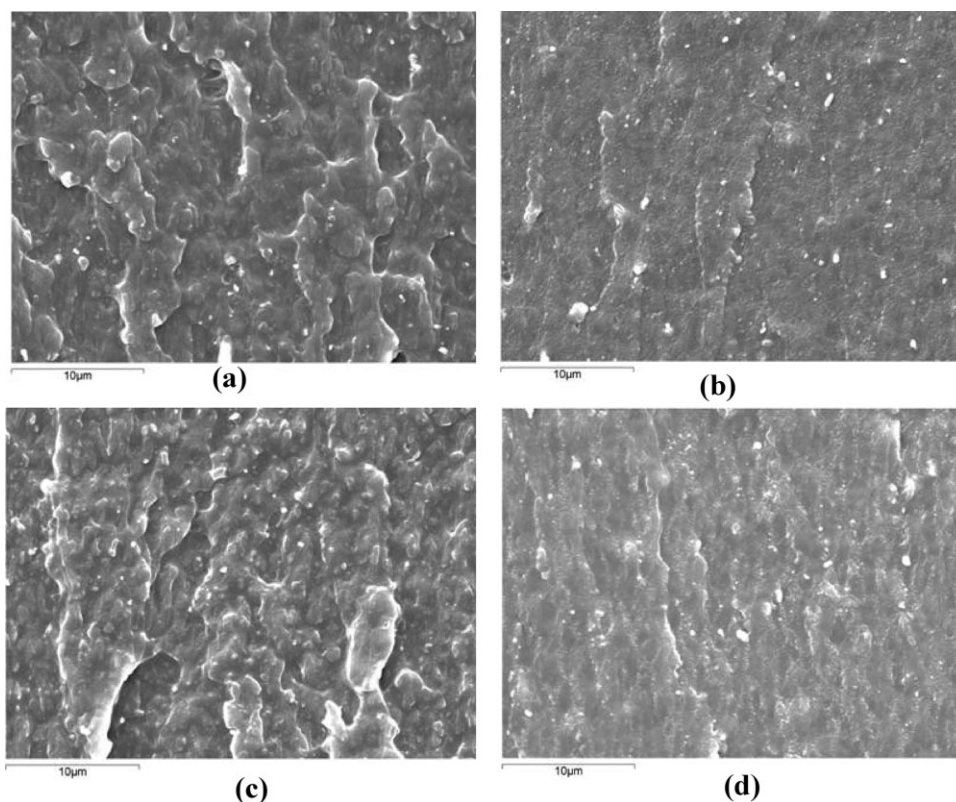


Figure 9 SEM micrographs of the tensile fractured surfaces of the gum blend vulcanizates at the magnification of $\times 3500$: (a) 70/30 NR/EPDM-simple; (b) 70/30 NR/EPDM-reactive; (c) 50/50 NR/EPDM-simple; and (d) 50/50 NR/EPDM-reactive.

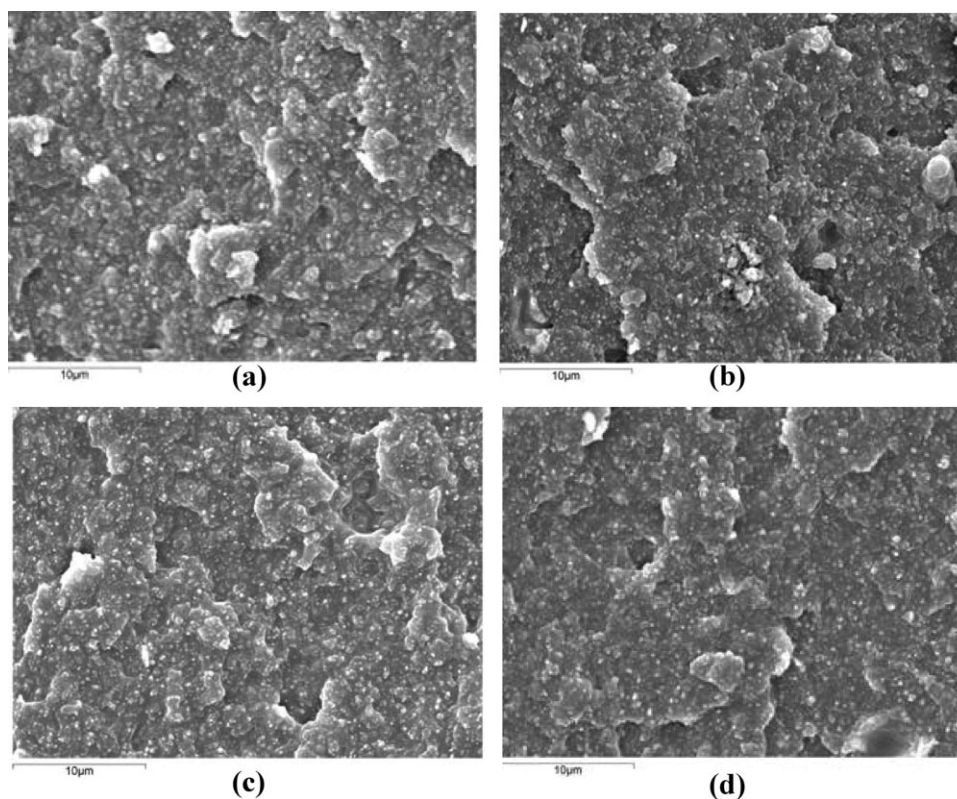


Figure 10 SEM micrographs of the tensile fractured surfaces of the blend vulcanizates filled with 50 phr of *N*-550 at the magnification of $\times 3500$: (a) 70/30 NR/EPDM-simple; (b) 70/30 NR/EPDM-reactive; (c) 50/50 NR/EPDM-simple; and (d) 50/50 NR/EPDM-reactive.

blends at both blend ratios, as illustrated in Figure 9, show a more smooth and homogeneous pattern in comparison with those of the simple blends. The better blend homogeneity in the reactive blends indicates the coherence of the two rubber phases, and hence, results in higher tensile strength or improved

mechanical properties. These SEM results of the tensile fracture surfaces are in good agreement with results reported by Ghosh et al.²² and Suma et al.¹⁶ for the same type of blends.

The fracture surfaces of the *N*-550-filled blends prepared by simple and reactive processing

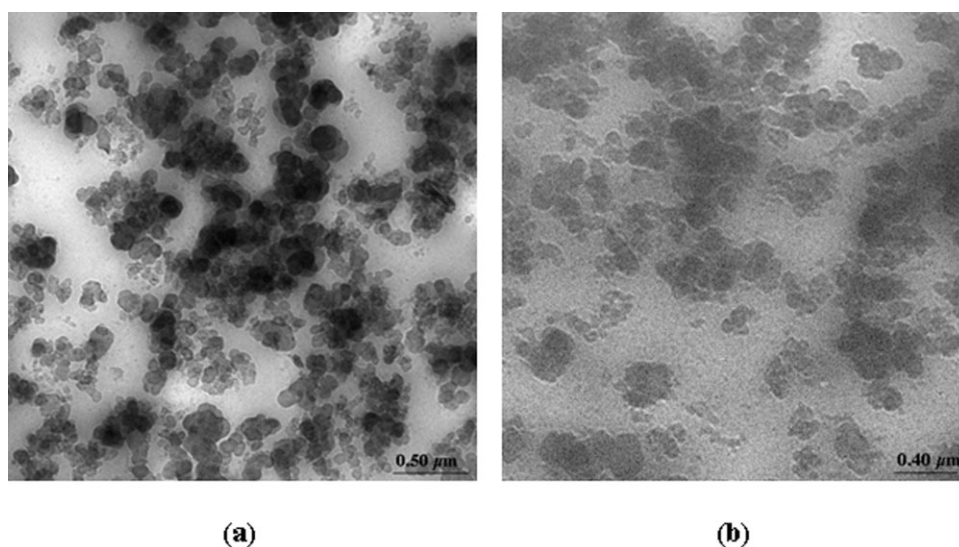


Figure 11 TEM micrographs of the blend vulcanizates filled with 50 phr of *N*-550 at the magnification of $\times 6000$: (a) 50/50 NR/EPDM-simple and (b) 50/50 NR/EPDM-reactive.

techniques at both blend ratios, as illustrated in Figure 10, show quite similar patterns. The better coherency of the rubber matrix and carbon black dispersion in the reactive blends were marginally visible in the SEM micrographs when compared with those of the simple blends. However, the TEM micrographs of the simple- and reactive-filled NR/EPDM blends at the blend ratio of 50/50, as shown in Figure 11, demonstrate that the carbon black was better distributed throughout the reactive blend in comparison with the simple blend. The dramatic improvement of tensile strength and elongation at break observed for the filled blends, as shown in Figures 7 and 8, should result from the improvement in both crosslink and filler distributions between the two rubber phases. For our NR/EPDM blends where both rubber phases showed very close glass transition temperature (T_g) and damping peaks are well overlapped, a quantitative analysis of filler distribution in the blends based on damping studies^{23,24} was not applicable.

CONCLUSIONS

The reactive processing technique applied to prepare the blends of NR/EPDM to reduce the migration of curatives and filler toward a higher reactive NR phase was proved to be very effective, as demonstrated by the improvement in ultimate tensile properties. For the gum blends at the blend ratio of 70/30, a significantly improved tensile strength of the reactive blends in comparison with that of the simple blends was observed when the CBS and MBTS were used. The increase of tensile strength in the reactive blends cured with the TBBS and MBT was marginal. The use of TBBS resulted in good mechanical properties in both simple and reactive 70/30 NR/EPDM blends. However, a dramatic improvement in ultimate tensile properties of the blends cured with the TBBS was observed when the NR/EPDM blend ratio was at 50/50. For the N-550-filled blends at the blend ratios of 70/30 and 50/50, the reactive-filled blends prepared under the optimized preheating times demonstrated superior tensile strength and elongation at break over the simple blends. The reactive technique used preheating of EPDM compound containing the entire amount of curatives to generate the rubber-bound intermediate prior to the crosslinking process. This results in less

curative migration toward the NR phase, more crosslinks in the EPDM phase, and less partition of carbon black in the highly unsaturated NR phase in the case of filled blends. The improvement of the blend vulcanizate properties therefore accounted for an improved crosslink and/or filler distribution between the two rubber phases, leading to more homogeneous blends as illustrated by the SEM and TEM micrographs.

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