

## Research Article

# Effects of Blend Ratio and SBR Type on Properties of Carbon Black-Filled and Silica-Filled SBR/BR Tire Tread Compounds

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This work aimed at investigating the effects of blend ratio between styrene butadiene rubber (SBR) and butadiene rubber (BR) and SBR type (E-SBR and S-SBR) on properties of SBR/BR tire tread compounds. Influences of these parameters on properties of the tread compounds reinforced by 80 parts per hundred rubber (phr) of carbon black (CB) and silica were also compared. Results reveal that hardness, strengths, and wet grip efficiency were impaired whereas rolling resistance was improved with increasing BR proportion. Surprisingly, the presence of BR imparted poorer abrasion resistance in most systems, except for the CB-filled E-SBR system in which an enhanced abrasion resistance was observed. Obviously, S-SBR gave superior properties (tire performance) compared to E-SBR, particularly obvious in the silica-filled system. Compared with CB, silica gave comparable strengths, better wet grip efficiency, and lower rolling resistance. Carbon black, however, offered greater abrasion resistance than silica.

## 1. Introduction

It is widely known that tire performance is justified by three main properties, that is, rolling resistance, wet grip efficiency, and abrasion resistance. Reducing rolling resistance of tire has gained much attention during the last two decades due to the increase demand for green transportation as tires are responsible for approximately 20–30% of vehicle's fuel consumption. Wet grip efficiency, or the handling ability of tire on wet road, is also of great importance as it is directly related to driving safety. Abrasion resistance is one of the key parameters to indicate the tire endurance. Many attempts have successively been made to investigate parameters affecting properties of tire tread compound with the goal of achieving improvements of both rolling resistance and wet grip efficiency of tire [1–12]. Previous works have shown that rolling resistance and wet grip efficiency are closely related to dynamic viscoelastic behavior of tread compound and can be represented by the ratio of loss modulus to storage modulus

( $\tan \delta$ ) at high temperature ( $\sim 60^\circ\text{C}$ ) and low temperature ( $\sim 0^\circ\text{C}$ ), respectively [8, 9, 13–17].

Carbon black (CB) has been widely used as the main reinforcing filler for tire tread compound because it offers greater processability with higher tire endurance, as compared with untreated silica. However, with the advent of silane coupling agent which can significantly improve silica dispersion and silica-rubber interaction, the use of silica technology has become popular for high performance tire nowadays. It has been reported that considerable reduction in rolling resistance can be achieved when carbon black is partially or fully replaced by silane-treated silica [16, 18]. In addition to silica technology, rubber blend technology is also of great interest for tire technologists, especially blends of styrene butadiene rubber (SBR) and butadiene rubber (BR) or natural rubber (NR) [3–5, 8, 10, 11, 18]. It has been reported that wear of a tire tread compound can be improved by the addition of BR [11]. Such finding,

TABLE 1: Typical properties of E-SBR and S-SBR employed in this study.

	S-SBR (6450SL)	E-SBR (1723)
Styrene content, %	34.6	23.4
Vinyl content, %	40.1	N/A
TDAE oil content, phr*	37.5	37.5
Mooney viscosity, MU**	53.2	49.1

\*Parts per hundred rubber. \*\*Mooney unit.

however, was obtained from the work carried out in CB-filled emulsion-polymerized SBR (E-SBR) system. Due to rapid progress in SBR synthesis, various grades of solution-polymerized SBR (S-SBR) are now commercially available. This offers tire technologists a better solution to achieve higher tire performance as S-SBR can be specially designed to give better controls of chain end structure, molecular weight distribution, and butadiene microstructure. It is therefore of great interest to investigate the effects of SBR/BR blend ratio and SBR type on properties of the tire tread compounds. In this work, investigation was made in two reinforcing systems for comparison purpose, that is, CB-filled system and silane-treated silica-filled system.

## 2. Experimental Section

**2.1. Materials.** Two types of commercial oil-extended SBR (E-SBR 1723 and S-SBR 6450SL) were produced by Kumho Petrochemical Co., Ltd., South Korea. Typical properties of both E-SBR and S-SBR are given in Table 1. Butadiene rubber (BR 01, ML1 + 4 @ 100°C = 45) was supplied by BST Elastomers Co., Ltd., Thailand. 2,2,4-Trimethyl-1,2-dihydroquinoline (TMQ), N-(1,3-dimethylbutyl)-N'-phenyl-p-phenylenediamine (6PPD), and N-tert-butyl-2-benzothiazole sulfenamide (TBBS) were obtained from Monflex Pte. Ltd., Singapore. Bis[3-(triethoxysilyl)propyl]tetrasulfide (TESPT) was purchased from Innova (Tianjin) Chemical Co., Ltd., China. Other additives were provided by suppliers in Thailand. Carbon black (N234; BET specific surface area = 126 m<sup>2</sup>/g) and silica (Tokusil 255; BET specific surface area = 166 m<sup>2</sup>/g) were obtained from Thai Carbon Black Public Co., Ltd. and OSC Siam Silica Co., Ltd., respectively. Treated distillate aromatic extract (TDAE) oil was supplied by PSP Specialties Co., Ltd. Stearic acid was obtained from Kij Paiboon Chemical Ltd., Part. Zinc oxide (ZnO) was manufactured by Thai-Lysaght Co., Ltd. Paraffin wax was supplied by Petch Thai Chemical Co., Ltd. Tetrabenzylthiuram disulfide (TBzTD) and sulfur were, respectively, supplied by Behn Meyer Chemicals (T) Co., Ltd., and Siam Chemicals Public Co., Ltd. All mixing ingredients were used as received.

**2.2. Preparation and Testing of Rubber Compounds.** Rubber compounds, with the formulations given in Table 2, were prepared using an internal mixer (Brabender-Plasticorder 350E, Germany) by a 3-step mixing technique. In the first step, the rubber pieces were mixed with all ingredients except curatives under the following conditions: fill factor = 0.75, initial chamber temperature = 60°C, rotor speed = 40

revolutions per minute (rpm), and mixing time = 10 minutes. The obtained compounds were subsequently mixed at high temperature (140°C) for 6 minutes to promote silanization reaction between silica and TESPT in the case of silica-filled system. The compounds were finally mixed with curatives in the third step using the same mixing conditions as in the first mixing step, except that the mixing time was reduced to 3 minutes. The compounds were sheeted on a two-roll mill (Labtech LRM150, Thailand) immediately after mixing and kept overnight prior to testing.

Mooney viscosity (ML1 + 4 @ 100°C) was measured by means of a Mooney viscometer (TechPro viscTECH+, USA) according to ISO 289-1. Optimum cure time ( $t_{c90}$ ) of the compounds was determined using a moving die rheometer (TechPro MD+, USA) at 160°C as per ISO 6502. Bound rubber content (BRC), an indicator of rubber-filler interaction, was measured by extracting the compounds with toluene at room temperature for 7 days. After filtration, the rubber-filler gel was completely dried in an oven at 70°C for 24 hours and weighed. The BRC was then calculated using

$$\% \text{ BRC} = \frac{W_{fg} - WF_f}{WF_p} \times 100, \quad (1)$$

where  $W_{fg}$  is the weight of rubber-filler gel,  $W$  is the test specimen weight,  $F_f$  is the weight fraction of filler, and  $F_p$  is the weight fraction of polymer in the rubber compound, respectively.

Magnitude of filler-filler interaction, widely known as Payne effect, was evaluated using a Rubber Process Analyzer (RPA 2000; Alpha Technologies, USA) under strain sweep test varying from 0.56% to 100% at 100°C and 1.7 Hz. Storage modulus ( $G'$ ) of the unvulcanized rubber compound was measured and the difference in  $G'$  at low (0.56%) and high (100%) strains ( $\Delta G'$ ) was used to represent the magnitude of filler-filler interaction. Hardness measurement was carried out using a Shore A durometer (Wallace, UK) in accordance with ISO 7619-1. Tensile and tear tests were performed on a universal testing machine (Instron 3366, USA) following ISO 37 (die type 1) and ISO 34 (method C), respectively. Abrasion resistance, represented in terms of volume loss, was determined using Akron-type abrasion tester (Gotech Model GT-7012-A, Taiwan) according to BS903 Part A9 (method B). Dynamic properties of the vulcanizates were measured in tension mode using a dynamic mechanical analyzer (Gabo, Eplexor 25N, Germany). The temperature sweep test was performed; that is, the temperature was scanned from -60 to 80°C at 2°C/min under static strain, dynamic strain, and frequency of 1%, 0.15%, and 10 Hz, respectively. Swelling ratio ( $Q$ ) was measured to indicate indirectly a magnitude of crosslink density. The test specimens were immersed in toluene at room temperature for 7 days. The swollen specimens were then blotted quickly with filter paper and finally weighed. Swelling ratio can be calculated from

$$Q = \frac{(W_2 - W_1)}{W_1}, \quad (2)$$

where  $W_1$  and  $W_2$  represent the weights of unswollen and swollen test specimens, respectively.

TABLE 2: Compound formulations.

Ingredient	SBR/BR blend ratio				
	100/0	90/10	80/20	70/30	0/100
SBR (6450SL or 1723)	137.5	123.75	110	96.25	0
BR	0	10	20	30	100
ZnO	3	3	3	3	3
Stearic acid	2	2	2	2	2
6PPD	1.5	1.5	1.5	1.5	1.5
TMQ	1	1	1	1	1
Paraffin wax	2	2	2	2	2
Filler (CB or Silica)	80	80	80	80	80
TESPT*	8	8	8	8	8
TDAE oil**	10	13.75	17.5	21.25	47.5
TBBS	1.2	1.2	1.2	1.2	1.2
TBzTD	0.2	0.2	0.2	0.2	0.2
Sulfur	2.2	2.2	2.2	2.2	2.2

\*TESPT was added only in silica-filled system. \*\*Total oil content was kept constant at 47.5 phr for all compounds.

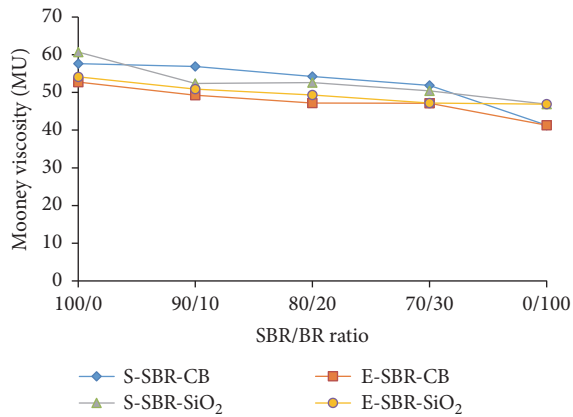


FIGURE 1: Mooney viscosity of the rubber compounds.

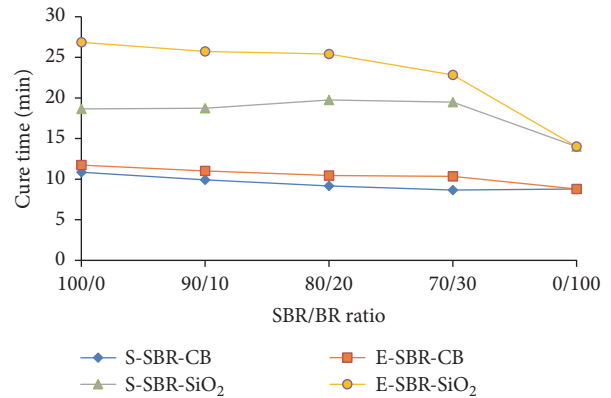


FIGURE 2: Cure time of the rubber compounds.

### 3. Results and Discussion

Mooney viscosities of the rubber compounds having different blend ratios are shown in Figure 1. Obviously, compound viscosity decreases with increasing BR content in the blends because BR used in this work has lower viscosity than E-SBR and S-SBR, respectively. At any given blend ratio, S-SBR/BR blends show slightly higher viscosity than E-SBR/BR blends which is attributed to the higher initial viscosity of S-SBR.

Results from MDR given in Figure 2 disclose the reduction of optimum cure time with increasing BR content in the blends. Explanation is given by the greater amount of double bonds in BR. Similar result has also been reported elsewhere [11]. Compared with E-SBR, S-SBR gives shorter cure time possibly due to the higher 1,2-vinyl content of S-SBR because it is accepted that vinyl configuration of butadiene is more reactive than other configurations [5]. Results also elucidate that silica-filled compounds have longer cure time than CB-filled compounds. The cure retardation of silica is not beyond expectation because silica has very low thermal conductivity

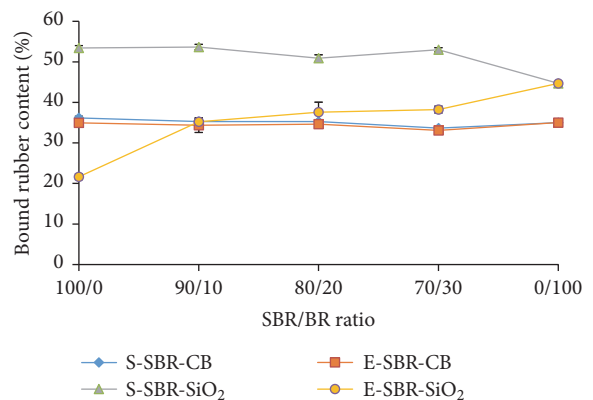


FIGURE 3: Bound rubber content (BRC) of the rubber compounds.

and the silanol groups on its surface can adsorb zinc complex and basic curatives necessary for sulfur vulcanization.

The relationship between BRC and blend ratio of the rubber compounds filled with CB and silica is depicted in Figure 3. For CB-filled system, BRC does not change

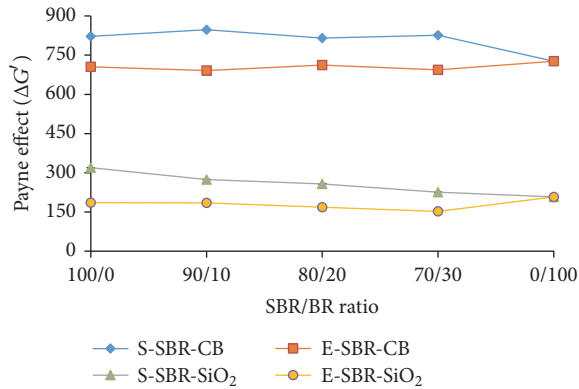


FIGURE 4: Payne effect of the rubber compounds.

significantly with increasing BR content. In addition, SBR type seems to have little effect on BRC in this reinforcing system. Since BRC is directly related to the magnitude of rubber-filler interaction, the results indicate that all rubber types employed in this study (E-SBR, S-SBR, and BR) have relatively good interaction with carbon black, possibly due to their matching polarity. Different results are found in silica-filled system in which S-SBR gives the highest magnitude of rubber-silica interaction (~52% BRC) whereas E-SBR gives the lowest (~22% BRC). The highest magnitude of rubber-silica interaction found in S-SBR arises from the higher reactivity of 1,2-vinyl groups in S-SBR towards silanization reaction. At sufficiently high mixing temperature, the alkoxy groups of TESPT could chemically react with hydroxyl groups on silica surface and the organogroups of TESPT could chemically react with the butadiene, particularly at the 1,2-vinyl groups in S-SBR leading to a greater magnitude of rubber-filler interaction. Despite the greater amount of double bonds, BR has slightly lower magnitude of rubber-silica interaction (~45% BRC) than S-SBR. This is attributed to the molecular configuration of BR as more than 97% of butadiene monomers are connected in cis-form which is less reactive than 1,2-vinyl form. Due to the lack of 1,2-vinyl form, the interaction between E-SBR and silica is relatively poor. Due to the dilution effect, S-SBR/BR blends exhibit a decreasing trend of BRC with increasing BR content whereas E-SBR/BR blends show the opposite trend.

Figure 4 illustrates the magnitude of filler-filler interaction or Payne effect ( $\Delta G'$ ) as a function of blend ratio of various rubber compounds. As Payne effects of BR and E-SBR are not significantly different both in CB-filled and in silica-filled systems, Payne effects of the E-SBR/BR blends therefore do not change with increasing BR content in both systems. However, for S-SBR which has higher Payne effect than BR, Payne effect tends to reduce with increasing BR content, particularly obvious in the silica-filled system. The results also clearly show that, with proper treatment of silica surface with TESPT, silica can give considerably lower filler-filler interaction than CB.

Mechanical properties such as hardness, 100% modulus ( $M_{100}$ ), tensile strength, and tear strength of the rubber vulcanizates are listed in Table 3. Regardless of the reinforcing

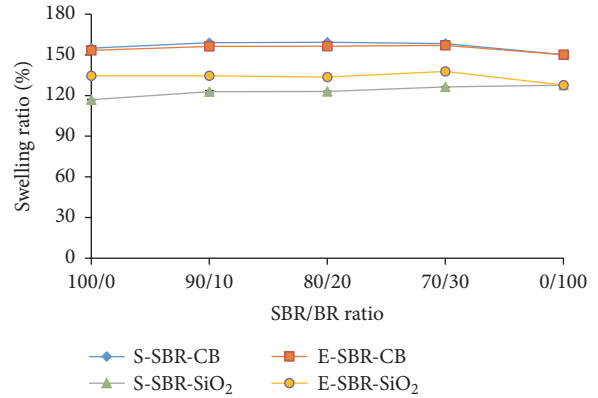


FIGURE 5: Dependence of swelling ratio on SBR/BR blend ratio.

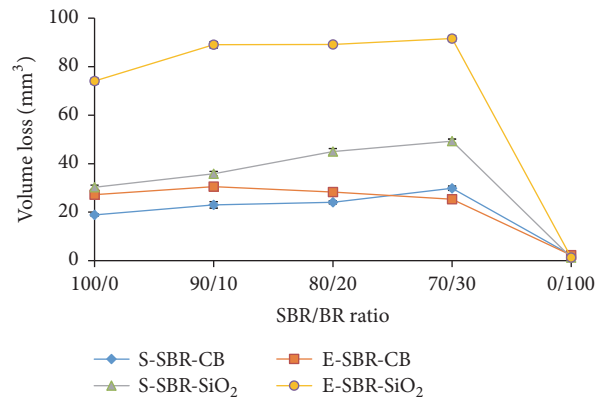


FIGURE 6: Effect of SBR/BR blend ratio on abrasion resistance.

system, BR has poorer mechanical properties than E-SBR and S-SBR and, thus, mechanical properties of the blends decrease continuously with increasing BR content. Similar observation was also reported elsewhere [11]. Obviously, at any given blend ratio, both E-SBR/BR and S-SBR/BR blends show comparable hardness, modulus, tensile strength, and tear strengths in the CB-filled system. This could be explained by the insignificant difference in rubber-filler interaction for this system (see also Figure 3). However, in the silica-filled system in which S-SBR possesses significantly greater rubber-filler interaction than E-SBR, S-SBR and its blends show significantly higher hardness and modulus than E-SBR counterparts. Interestingly, both tensile strength and tear strength of silica-filled E-SBR/BR and S-SBR/BR blends are not significantly different when compared at the same blend ratio. The results imply that magnitude of rubber-filler interaction strongly affects modulus and hardness and has minor effect on strengths of the rubber vulcanizates in this work. The results also reveal that the silica-filled system gives higher modulus than the CB-filled system. The combined effects of greater rubber-filler interaction and higher crosslink density caused by the released sulfur from TESPT, as indicated by the lower swelling ratio in Figure 5, are used to explain the results.

Figure 6 depicts the dependence of abrasion resistance on rubber blend ratio and SBR type. As expected, BR shows

TABLE 3: Hardness, modulus, tensile strength and tear strength of the rubber vulcanizates.

Blend ratio	CB-filled system				Silica-filled system			
	Hardness (Shore A)	Tensile strength (MPa)	$M_{100}$ (MPa)	Tear strength (N/mm)	Hardness (Shore A)	Tensile strength (MPa)	$M_{100}$ (MPa)	Tear strength (N/mm)
S-SBR/BR								
100/0	61.2 ± 1.1	16.9 ± 0.6	2.09 ± 0.04	103.5 ± 1.3	64.9 ± 0.5	18.7 ± 1.3	3.36 ± 0.01	115.5 ± 4.7
90/10	59.9 ± 0.4	16.6 ± 0.6	1.78 ± 0.04	101.8 ± 1.2	62.4 ± 0.4	17.4 ± 0.9	2.63 ± 0.09	113.8 ± 3.1
80/20	60.0 ± 0.4	15.7 ± 0.3	1.77 ± 0.02	99.4 ± 1.6	60.9 ± 0.4	17.0 ± 0.8	2.46 ± 0.04	109.9 ± 2.9
70/30	59.9 ± 0.2	15.9 ± 0.4	1.80 ± 0.02	94.8 ± 1.9	61.1 ± 0.4	16.1 ± 0.6	2.38 ± 0.03	108.4 ± 4.6
E-SBR/BR								
100/0	62.1 ± 0.2	18.6 ± 0.4	1.87 ± 0.03	106.2 ± 3.1	60.6 ± 0.2	19.8 ± 0.8	2.31 ± 0.06	122.3 ± 2.2
90/10	60.0 ± 0.4	17.5 ± 0.8	1.76 ± 0.03	103.9 ± 4.1	58.1 ± 0.5	18.8 ± 0.6	2.42 ± 0.06	115.6 ± 3.7
80/20	60.7 ± 0.7	17.5 ± 0.5	1.72 ± 0.03	99.9 ± 1.3	59.3 ± 0.6	17.0 ± 0.6	2.45 ± 0.04	112.2 ± 3.8
70/30	60.5 ± 0.4	16.5 ± 0.3	1.70 ± 0.03	90.3 ± 2.9	58.1 ± 0.7	16.8 ± 0.7	2.17 ± 0.02	99.0 ± 4.5
Pure BR								
0/100	58.7 ± 0.8	12.3 ± 0.4	1.50 ± 0.04	52.6 ± 2.2	57.6 ± 0.7	13.2 ± 0.2	1.73 ± 0.03	78.8 ± 3.2

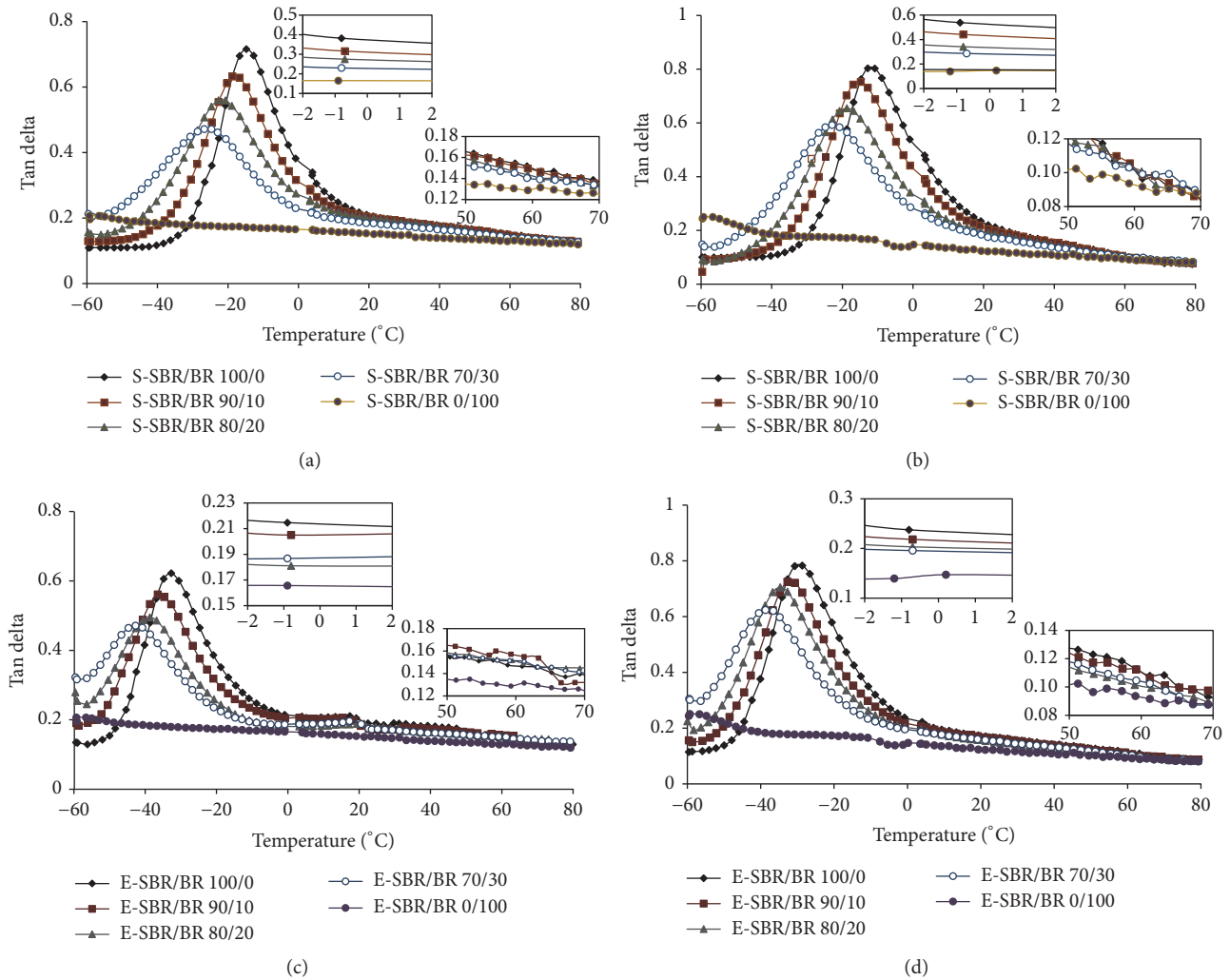


FIGURE 7: Dependence of  $\tan \delta$  on SBR/BR blend ratio of the rubber vulcanizates; (a) CB-filled S-SBR/BR, (b) SiO<sub>2</sub>-filled S-SBR/BR, (c) CB-filled E-SBR/BR, and (d) SiO<sub>2</sub>-filled E-SBR/BR.

the highest abrasion resistance as confirmed by its lowest volume loss both in CB-filled and in silica-filled systems. It is also found that S-SBR gives superior abrasion resistance than E-SBR which is particularly obvious in the silica-filled system due to the greater rubber-filler interaction and higher crosslink density as previously discussed. Surprisingly, the improvement of abrasion resistance with increasing BR content is found only in the CB-filled E-SBR system which is in good accordance with the work previously reported by Karak and Gupta [11]. However, the unexpected deterioration of abrasion resistance with increasing BR content is found in the other systems. Such deterioration could be explained by the reductions of hardness and tear strength with increasing BR content which override the dilution effect. Obviously, at any given blend ratio, the CB-filled system gives noticeably higher abrasion resistance than the silica-filled system. The stronger filler-filler network is thought to be the reason for such finding.

The curves of  $\tan \delta$  as a function of temperature of the rubber vulcanizates are shown in Figure 7. Generally, glass

transition temperature ( $T_g$ ) is defined as the temperature at  $\tan \delta$  peak. As  $T_g$  of BR is below  $-60^\circ\text{C}$ , only  $T_g$  values of E-SBR and S-SBR are observed at  $-32.7^\circ\text{C}$  and  $-15^\circ\text{C}$ , respectively. The higher  $T_g$  of S-SBR is attributed to the greater styrene and 1,2-vinyl contents in association with its higher crosslink density. Apparently,  $T_g$  values of E-SBR/BR and S-SBR/BR blends decrease continuously with increasing BR content which could be attributed by the good compatibility between SBR and BR. Due to the dilution effect, the magnitude of  $\tan \delta$  peak at  $T_g$  of SBR decreases with increasing BR content.

In tire industry, the values of  $\tan \delta$  at  $0^\circ\text{C}$  and  $60^\circ\text{C}$  are, respectively, used to indicate wet grip efficiency and rolling resistance of a tread compound. Since the test temperature employed in this study is far above  $T_g$  of BR, thus, BR molecules are in the rubbery zone throughout the test temperature range leading to the relatively low  $\tan \delta$  values at both  $0^\circ\text{C}$  and  $60^\circ\text{C}$ . The BR compounds therefore show the poorest wet grip efficiency with the lowest rolling resistance

TABLE 4: Values of  $\tan \delta$  at 0°C and 60°C of the rubber vulcanizates.

Blend ratio	$\tan \delta$			
	Carbon black		Silica	
	0°C	60°C	0°C	60°C
S-SBR/BR				
100/0	0.381	0.153	0.538	0.105
90/10	0.316	0.149	0.441	0.105
80/20	0.275	0.143	0.341	0.103
70/30	0.230	0.140	0.286	0.103
E-SBR/BR				
100/0	0.206	0.160	0.237	0.112
90/10	0.205	0.157	0.218	0.110
80/20	0.187	0.151	0.203	0.106
70/30	0.181	0.151	0.196	0.105
Pure BR				
0/100	0.165	0.130	0.147	0.093

(implying the best fuel consumption efficiency). Because S-SBR has significantly higher  $T_g$  than E-SBR and, at 0°C,  $\tan \delta$  value of S-SBR is at the peak shoulder whereas that of E-SBR is at the base of the curves, S-SBR therefore gives remarkably higher wet grip efficiency than E-SBR. Table 4 summarizes the  $\tan \delta$  values at 0°C and 60°C of the blends as extracted from Figure 7. As expected, the results reveal the reduction of wet grip efficiency ( $\tan \delta$  at 0°C) of the blends with increasing BR content, regardless of SBR or filler types. Also, shown in Table 4, silica gives apparently better wet grip efficiency than CB which is in good accordance with other works [8, 18, 19].

Results in Table 4 also show that rolling resistance of SBR/BR blends decreases with the increase of BR content in both CB-filled and silica-filled systems. At any given blend ratio, S-SBR/BR exhibits slightly lower rolling resistance than E-SBR/BR, possibly due to the greater rubber-filler interaction and higher crosslink density. Evidently, silica gives significantly lower rolling resistance than CB. Again, the greater rubber-filler interaction, higher crosslink density, and lower filler-filler interaction are the possible explanations.

#### 4. Conclusions

Increasing BR content in SBR/BR blends not only causes impairments of most mechanical properties such as strengths, modulus, and hardness but also leads to the reduced wet grip efficiency. The increase of BR content, however, provides the tread with improved rolling resistance and thus better fuel consumption efficiency. Surprisingly, improvement of abrasion resistance with increasing BR content is found only in CB-filled E-SBR compounds whereas opposite result is observed in the others. Compared with E-SBR, S-SBR exhibits comparable tensile and tear strengths with greatly improved tire performance, that is, higher wet grip efficiency, lower rolling resistance, and greater abrasion resistance. The use of silica in place of CB not only noticeably improves modulus but also results in a greater wet grip efficiency and lower rolling resistance. This phenomenon is more pronounced in S-SBR and its blends because S-SBR

can form stronger interaction with silica via TESPT as compared with E-SBR. Nevertheless, the use of CB offers longer service life of tread because CB gives greater abrasion resistance than silica. The results imply that, to achieve balanced tire performance, S-SBR should be selected and the reinforcement by CB/silica hybrid filler is recommended.

#### Conflicts of Interest

The authors declare that they have no conflicts of interest.

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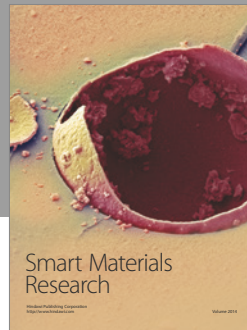
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