

Enhancement of Rubber–Carbon Black Interaction by Amine-Based Modifiers and their Effect on Viscoelastic and Mechanical Properties

Kannika Hatthapanit,^{1,2} Pongdhorn Sae-Oui,² Narongrit Sombatsompop,³ Chakrit Sirisinha^{1,4}

¹Department of Chemistry and Center of Excellence for Innovation in Chemistry, Faculty of Science, Mahidol University, Bangkok, Thailand

²National Metal and Materials Technology Center, Thailand Science Park, Klong Luang, Pathumthani, Thailand

³Polymer Processing and Flow Group, School of Energy, Environment and Materials, King Mongkut's University of Technology Thonburi, Bangkok, Thailand

⁴Research and Development Centre for Thai Rubber Industry, Faculty of Science, Mahidol University, Salaya, Nakhon Pathom, Thailand

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ABSTRACT: Improvement in rubber–filler interaction is desirable for rubber technologists due to its influence on numerous properties of rubber compounds and vulcanizates. In practice, there are coupling agents commercially available for the improvement of silica–rubber interaction. Surprisingly, only a limited number of works have been focused on interaction enhancement between carbon black (CB) and rubber. Thus, in the research presented in this article, attempts to improve interaction between ethylene–propylene rubber (EPM) and carbon black (CB) have been made by the use of either *p*-phenylene diamine (*p*-PDA) or *N*-*tert*-butyl-2-benzothiazole sulfenamide (TBBS) as an interaction modifier. Bound rubber content (BRC), used as an indicator for rubber–filler interaction and viscoelastic behavior of CB masterbatches and CB-filled EPM compounds were investigated and correlated. Results from the measurement of BRC in the CB masterbatches revealed

that *p*-PDA was more effective in the enhancement of rubber–CB interaction than TBBS. Such improved interaction led to a decrease in magnitude of CB percolation (Payne effect). In respect of viscoelastic behavior, the interaction modifiers affected G' only in the small strain region ($<1\%$ strain) by slightly raising the value of G' . However, as strain was increased ($\geq 1\%$), G' for all compounds was coincident implying a disruption of weak interaction between CB and rubber. In the case of EPM vulcanizates, *p*-PDA yielded greater enhancement in mechanical properties than TBBS. The results of BRC, viscoelastic behavior, and mechanical properties were apparently in good agreement. © 2012 Wiley Periodicals, Inc. *J Appl Polym Sci* 000: 000–000, 2012

Key words: rubber–filler interaction; carbon black; bound rubber; viscoelastic behavior; EPM

INTRODUCTION

Dynamic mechanical properties of rubber are known to be important in numerous engineering products including tires and vibration isolators. To achieve good dynamic properties with low hysteresis, a high-elastic contribution is required and viscoelastic effects had to be minimized.¹ Practically, the reinforcing fillers particularly carbon black (CB) and precipitated silica (PSi) are incorporated into rubber to enhance mechanical properties of the vulcanizates. However, with poor rubber–filler interaction, molecular slippage of the rubber molecules can take place at the filler surfaces leading to a viscous contribution and thus to heat build-up (HBU).² Fortunately, in PSi-filled systems, enhancement of rubber–PSi inter-

action is practically achieved by the use of a silane coupling agent (SCA). Typically, the SCA is composed of two functionally active end groups, i.e., the readily hydrolysable alkoxy group for reacting chemically with the silanol groups on silica surfaces and the organo-functional group for being compatible with rubbers. By this means, the strong chemical linkages between silica and rubber are developed, resulting in a decrease in molecular slippage at the silica surfaces. For CB, its surface reactivity comes mainly from the oxygen-contained functional groups including ketone, aldehyde, and hydroxyl, which provide interaction between CB and the rubber matrix.³ In general, the interaction between CB and rubber takes place through Van der Waals forces, which are weaker than the covalent bonds found in silane-treated silica systems. Molecular slippage of rubber molecules at CB surfaces can therefore easily take place, especially at high strains. The energy dissipation process occurring from such slippage leads to the development of heat under cyclic deformation,

Correspondence to: C. Sirisinha (sccsr@mahidol.ac.th).

which is undesirable. This is usually termed heat build-up (HBU). Attempts to improve interaction between CB and rubber have been carried out using various approaches such as modification of CB surface⁴ or rubber matrix⁵ and synthesis of coupling agent.^{6,7} Some published work has reported that CB dispersion can be enhanced by grafting polymers onto CB surfaces via polycondensation reaction to produce functionalized CB.^{8–13} In addition, it has been shown that the CB surface oxidation by oxygen plasma or oxidizing agents to generate the oxygen-containing functional groups on CB surfaces promotes their reactivity.¹⁴ However, these techniques are not yet practical due to the complication in treatment conditions and therefore the solid-state modification of CB surfaces or *in situ* method has gained interest recently. For example, CB modification based on the radicals generated from the rubber matrix as a result of mechanical force has been reported.^{15–17} In addition, some amine-based reagents, including *p*-aminobenzenesulfonyl azide and dinitrosodiamine, have been found to provide CB–rubber interaction enhancement via a coupling reaction.^{6,18} Thus, the aim of the research described in this article is to improve interaction between ethylene–propylene rubber and carbon black by the use of maleic anhydride grafted liquid polybutadiene (Ricon 130MA8) in association with either *p*-phenylene diamine (*p*-PDA) or *N*-*tert*-butyl-2-benzothiazole sulfenamide (TBBS). It is anticipated that the rubber–carbon black interaction to be developed in the present work is the result of interactions among amine groups of *p*-PDA or TBBS, active functional group on carbon black surfaces (e.g., lactone, aldehyde, and carboxyl groups), maleic anhydride (MA) groups of Ricon130MA8 and an EPM matrix. Bound rubber content (BRC, as an indication of rubber–filler interaction) and viscoelastic behavior of rubber compounds were investigated and correlated.

EXPERIMENTAL

Materials

All mixing ingredients were used as-received. Ethylene–propylene rubber (EPM, Keltan 520 with 60% ethylene content) was supplied by DSM elastomers Asia, The East Asiatic (Thailand), Thailand. The N330 carbon black was manufactured from Thai Carbon Products, Thailand. Functionalized liquid polybutadiene (Ricon 130MA8) was purchased from Chemical Innovation, Thailand. As claimed by the manufacturer, there is 8% maleic anhydride adducted, 20–35% 1, 2 vinyl content and 3,100 g/mol number-average molecular weight. The *N*-*tert*-butyl-2-benzothiazole sulfenamide (Santocure-TBBS) was purchased from Reliance Technochem (Flexsys), Thailand. The *p*-phenylene diamine (*p*-PDA) was purchased from

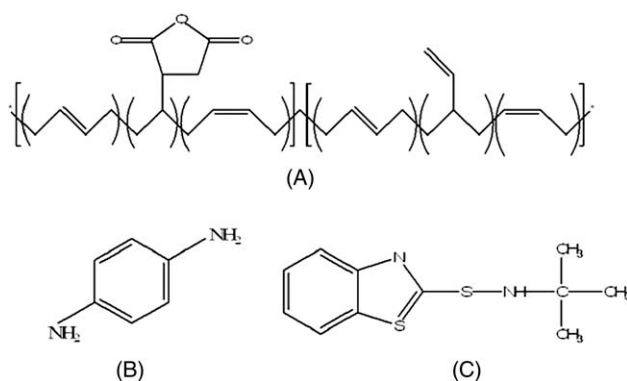


Figure 1 Chemical structures of: (a) Ricon130MA8, (b) *p*-PDA, and (c) TBBS.

Sigma-Aldrich, India. Dicumyl peroxide 40% dispersed in kaolin (Luperox DC40KE) was received from Akema, India. Chemical structures of Ricon130MA8, *p*-PDA, and TBBS are shown in Figure 1.

Compound preparation

The preparation procedure of EPM compounds was carried out using a two-stage mixing process and compound formulations are illustrated in Table I. Mixing was performed using a laboratory-scale internal mixer (Brabender Plasticorder, Germany) at a rotor speed, set temperature and fill factor of 40 rpm, 140°C and 0.7, respectively. In the first stage, 100 phr CB was mixed with pre-masticated EPM for 4 min followed by the addition of 50 phr Ricon 130MA8. The mixing was continued for 5 min and then various constituents, ranging from 0 to 0.20 phr, of either *p*-PDA or TBBS were added and mixed further until 15 min of mixing time had elapsed. The mix was discharged from the mixing chamber and denoted as “CB masterbatch (CB-M/B).” In the second stage of mixing, the CB-M/B was diluted with raw EPM to achieve final CB loading of 45 phr. The mixing was performed at a mixing temperature of 60°C for 2 min. Finally, Luperox DC40KE was added into the compound 3 min prior to being discharged. The mix was then sheeted on a two-roll mill (Collin W100T, Germany) and denoted as “EPM compound.” It had to be noted that during the experiment, a wide range of *p*-PDA loadings were used, but the maximum loading of *p*-PDA to be used in this work was 0.2 phr. The EPM compounds with higher *p*-PDA loadings above 0.2 phr were carried out, but found to have encountered cure suppression phenomenon, which was caused by the excessive amine group acting as a radical scavenger during the peroxide vulcanization process.¹⁹

Testing of rubber compounds

The determination of BRC was based on the dissolution technique. The preweighed rubber compounds

TABLE I
Compound Formulations

Materials	Loading (phr)
Carbon black masterbatches (CB-M/B)	
EPM (Keltan 520)	100
CB N330	100
Ricon 130MA8	50
<i>p</i> -PDA	0–0.20
TBBS	0–0.20
EPM compounds	
EPM (Keltan 520)	55
CB-M/B	112.5
Luperox DC40KE	5

were immersed in toluene, an appropriate solvent for EPM, to dissolve the unbound rubber. The dissolution was carried out at the temperature of 85°C for 1 day. The insoluble part was then filtered and dried overnight at 70°C prior to weighing. Thereafter, the rubber content in the insoluble part was measured using a thermogravimetric analyzer (TGA, Mettler SDTA851) over a temperature range of 40–600°C at a heating rate of 20°C/min. The value of BRC was calculated using eq. (1).²⁰

$$\text{BRC}(\%) = \frac{W_d - F}{R} \times 100 \quad (1)$$

where W_d was the weight of the dried gel.

F was the weight of the filler in the gel (same as weight of the filler in the original sample) and R was the weight of the polymer in the original sample.

Viscoelastic behavior of compounds was monitored by the use of an oscillatory rheometer, namely, Rubber Process Analyzer (RPA2000, Alpha Technology). Strain and frequency sweep tests were conducted by varying strain from 0.5 to 1000% at a test frequency of 10 rad/s and by varying frequency from 0.1 to 100 rad/s at a given strain of 1% (within the linear viscoelastic region), respectively. The dynamic storage moduli (G') as functions of strain and angular frequency at 100°C were then recorded. The difference in storage moduli at low and high strains ($\Delta G'$), termed the "Payne effect," is used to represent the degree of filler–filler interaction.²¹

Cure characteristics were determined using a moving die rheometer (TechPro RheoTech MD+) at 160°C. The torque difference ($M_H - M_L$) was used to represent the crosslink density of the vulcanizates.²²

Mechanical properties of rubber vulcanizates measured in this work include hardness and tensile properties, and dynamic loss. The tensile test was conducted using a universal testing machine (Instron model 5566, USA). Tensile strength, elongation at break, and modulus at 100% strain (M_{100}) were determined as per ASTM D412. The hardness was

determined according to ASTM D2240 using a Wallace Shore A durometer (Wallace Cogenix, UK).

RESULTS AND DISCUSSION

Bound rubber content

Typically, the degree of interaction between filler and rubber could be quantified by a measure of BRC.^{20,23} Higher BRC implies the greater interaction between rubber (i.e., EPM in this case) and CB. Figure 2 reveals that, with increasing *p*-PDA based modifier, BRC of the CB-M/B tends to increase continuously. The results imply that the development of rubber–CB interaction could be enhanced by the addition of *p*-PDA. Probably, the amine groups of *p*-PDA could act as connections between maleic anhydride (MA) group of Ricon130MA8 and the functional groups (e.g., lactone, carboxylic, or hydroxyl groups) on the carbon black surface. Since the number of active groups on a CB surface is reported to be relatively small (e.g., 1–2% active oxygen in furnace blacks),²⁴ the magnitude of a BRC increment is not very high. Unlike *p*-PDA, TBBS does not appear to significantly alter BRC. The greater rubber–filler enhancement efficiency of *p*-PDA might be caused by the relatively low magnitude of steric hindrance leading to the relatively high opportunity for amine groups to react with active sites on CB surfaces (e.g., lactone, aldehyde, and carboxyl groups) and MA groups of Ricon 130MA8.

Figure 3 represents the BRC results of the EPM compounds prepared by diluting the modified CB-M/B with virgin EPM. It is evident that, at any given modifier loading, BRC of the EPM compounds is slightly higher than that of the CB-M/B. Such additional interaction found in the EPM compounds might be attributed to the free radicals generated during the mixing process. Also, the interaction developed from such radicals in a large EPM matrix might govern the total magnitude of bound rubber formation in the EPM compounds. It is evident that

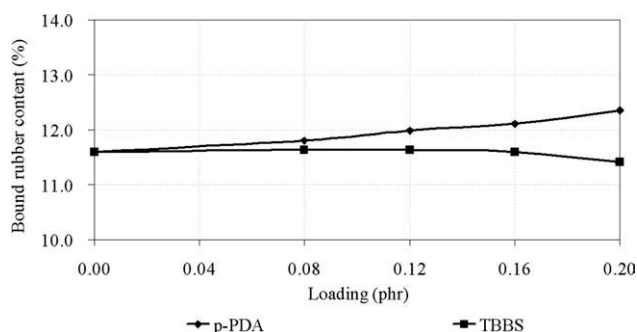


Figure 2 Bound rubber content (BRC) of carbon black masterbatches (CB-M/B) modified with various loadings of *p*-PDA and TBBS.

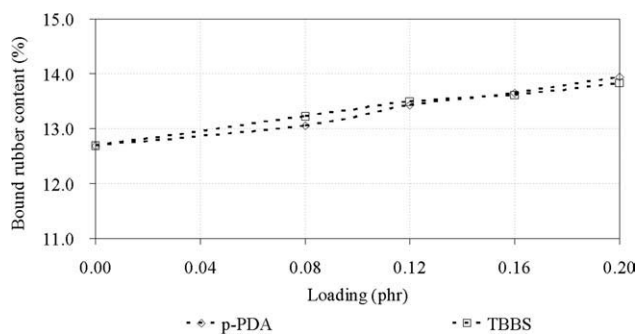


Figure 3 Bound rubber content (BRC) of EPM compounds modified with various loadings of *p*-PDA and TBBS.

the BRC of the EPM compounds increases progressively with increasing loading of the modifiers, regardless of modifier type.

Viscoelastic behavior

Figures 4(a) and 4(b) show viscoelastic behavior of the CB-M/B modified with various loadings of *p*-PDA and TBBS, respectively. It is evident that there is no torque rise with time indicating no sign of crosslink development by the incorporation of *p*-PDA or TBBS into the CB-M/B. In other words, the increase in BRC by the use of interaction modifier as reported previously appears not to be caused by the scorch phenomenon of the CB-M/B. However, it could be that the thermal degradation gives

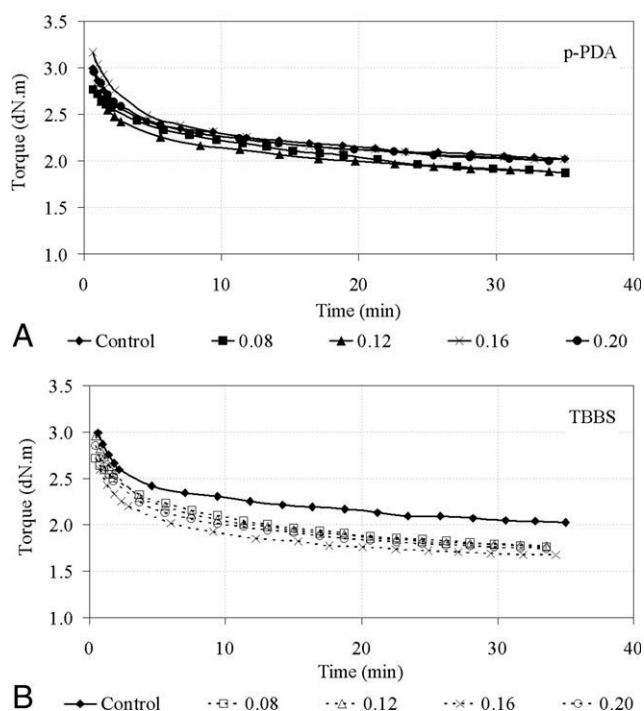


Figure 4 Viscoelastic behavior of carbon black masterbatches (CB-M/B) modified with various loadings of: (a) *p*-PDA and (b) TBBS.

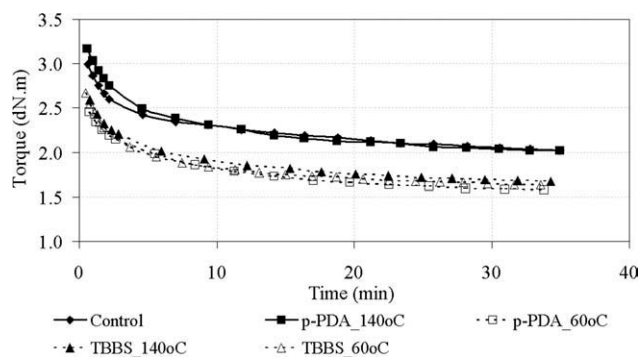


Figure 5 Viscoelastic behavior of carbon black masterbatches (CB-M/B) modified with 0.16 phr of *p*-PDA and TBBS prepared at 140 and 60°C.

rise to the misleading torque results observed. To clarify this point, the modification process temperature was reduced from 140 to 60°C, so that the thermal degradation effect could be minimized. According to the Arrhenius concept, the systems with no thermal degradation should demonstrate relatively high bulk viscosity due to the minimal magnitude of thermal degradation. As shown in Figure 5, the system prepared at the high temperature of 140°C still revealed higher torque than that at 60°C, supporting the development of rubber–filler interaction at high temperature.

Figure 6 exhibits the results of strain-dependent modulus ($\Delta G'$) which is an indication of the Payne effect of the CB-M/B modified with different modifiers. As reported elsewhere, the decrease in magnitude of $\Delta G'$ is used as an implication of reduction in filler–filler interaction and thus CB percolation. Such $\Delta G'$ reduction would then lead to an improvement in filler dispersion degree.^{21,25} Thus, as evidenced from Figure 6, both modifiers gave decreasing in $\Delta G'$ of the CB-M/B implying the enhancement in the degree of filler dispersion. The magnitude of $\Delta G'$ tended to decrease with increasing modifier loading and approached the minimum at 0.12 and 0.16 phr of *p*-PDA and TBBS, respectively. The results imply

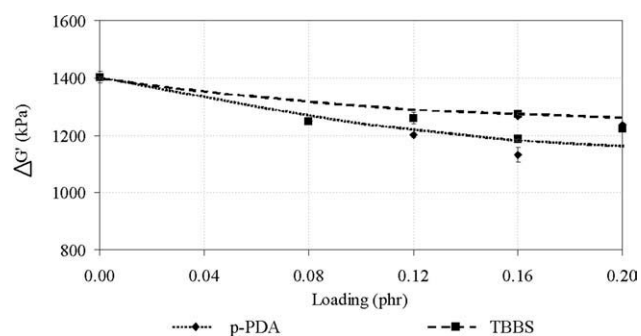


Figure 6 Magnitude of the Payne effect of carbon black masterbatches (CB-M/B) influenced by either *p*-PDA or TBBS.

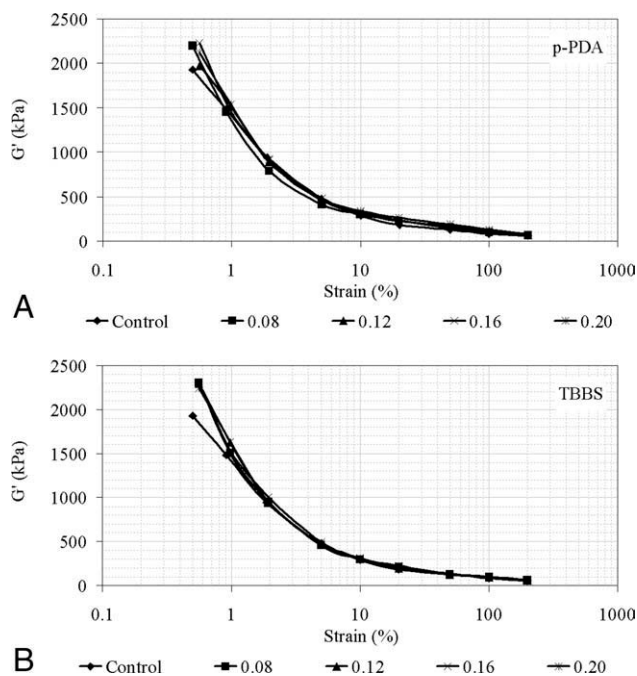


Figure 7 Strain sweep test results of the EPM compounds with various loadings of (a) *p*-PDA or (b) TBBS as interaction modifier.

that the presence of *p*-PDA or TBBS in the carbon black-filled EPM could enhance the degree of CB dispersion to some extent. However, in the case of the EPM compounds where the modified CB-M/B was diluted by raw EPM, it is evident from Figure 7 that only a slight increase in G' was observed, specifically in the small strain region ($<1\%$). Beyond this point, G' for all compounds is coincident. The drop in G' at high strain is probably caused by the disruption of filler network and/or filler-rubber interaction.^{26,27} The similarity in G' results of the EPM compounds regardless of modifier type is in good agreement with the BRC results as shown previously in Figure 3.

Figure 8 illustrates cure behavior of the EPM compounds with various contents of *p*-PDA or TBBS as interaction modifiers. It appears that no significant change in cure behavior was observed in the presence of *p*-PDA and TBBS. An exception was found in the cure curve of the compound having 0.2 phr of *p*-PDA as a modifier, in which the torque rise is greater than the others. In fact, at high loading of *p*-PDA, larger amounts of amine groups are available to react with MA group in Ricon130MA8. It is widely accepted that the MA group is a very reactive group which could react rapidly with peroxide^{28,29} and thus the presence of MA in rubber molecule could retard the peroxide vulcanization. At 0.2 phr of *p*-PDA, it is thought that all of the MA groups in Ricon130MA8 might fully be reacted with amine groups. More free radicals from peroxide

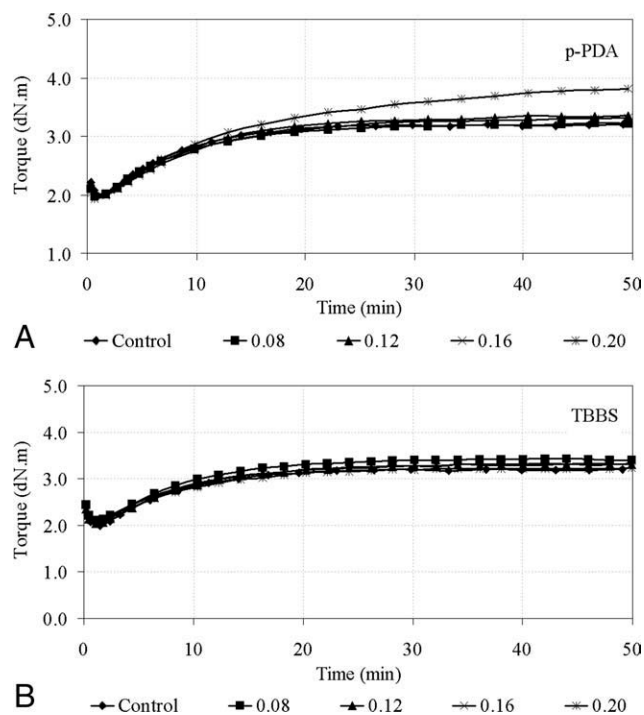


Figure 8 Cure behavior of EPM compounds modified with various loadings of: (a) *p*-PDA and (b) TBBS.

were therefore available for crosslink reaction particularly in the Ricon130MA8 phase leading to greater state-of-cure. At low loadings of *p*-PDA, some MA groups were still available in the Ricon130MA8 resulting in a relatively low degree of crosslink density in the Ricon130MA8 phase. The cure curve characteristics were thus mainly controlled by the crosslink of the EPM phase. No significant change in cure curve characteristics was therefore observed. A similar explanation is also applied to the TBBS. As TBBS has only one groups of amine per molecule, it is believed that 0.2 phr of TBBS was not enough to fully remove the MA groups in Ricon130MA8 phase. No significant difference in cure curve was therefore observed.

To support the proposed explanation of cure behavior affected by interaction modifiers, the cure characteristics of the following systems were

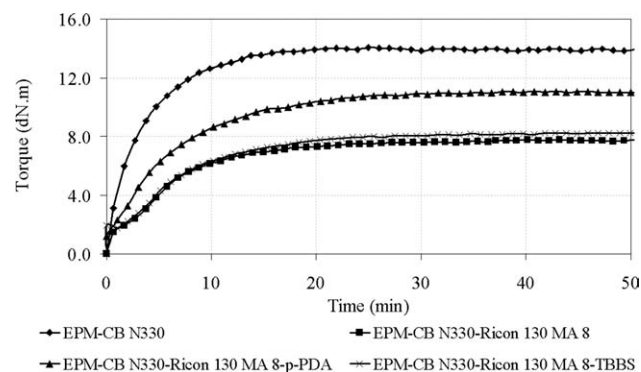


Figure 9 Cure behavior affected by interaction modifiers.

TABLE II
Comparison of Mechanical Properties of EPM Vulcanizates with Various Loadings of *p*-PDA or TBBS as Interaction Modifiers

	Loading (phr)	TS (MPa)	EB (%)	M_{100} (MPa)	Hardness (Sh A)
Control	0.00	3.73 ± 0.64	342.5 ± 12.9	1.64 ± 0.01	62.1 ± 1.0
<i>p</i> -PDA	0.08	5.36 ± 0.27	411.9 ± 15.5	1.61 ± 0.02	61.6 ± 0.4
	0.12	4.99 ± 0.05	372.3 ± 8.8	1.71 ± 0.02	62.8 ± 0.5
	0.16	6.31 ± 0.14	429.1 ± 11.7	1.63 ± 0.01	63.1 ± 0.7
	0.20	7.17 ± 0.09	429.0 ± 10.9	1.73 ± 0.01	65.2 ± 0.7
TBBS	0.08	4.99 ± 0.47	380.7 ± 12.7	1.61 ± 0.01	63.6 ± 0.9
	0.12	5.00 ± 0.44	380.5 ± 19.7	1.57 ± 0.01	62.3 ± 0.5
	0.16	5.04 ± 0.15	393.6 ± 9.7	1.57 ± 0.02	62.3 ± 0.7
	0.20	4.88 ± 0.20	403.9 ± 10.0	1.48 ± 0.03	62.7 ± 1.3

measured: System (I) containing EPM with unmodified CB; System (II) having EPM with CB and Ricon130MA8; System (III) containing EPM with CB, Ricon130MA8 and *p*-PDA; System (IV) having EPM with CB, Ricon130MA8, and TBBS. As illustrated in Figure 9, the System (I) as a control system exhibited the highest state-of-cure. By the presence of Ricon130MA8, the state-of-cure observed in System (II) reduced drastically, implying the cure retardation phenomenon of EPM matrix by the Ricon130MA8. The incorporation of *p*-PDA could reduce the magnitude of the cure retardation phenomenon (see System III). Unlike *p*-PDA, the TBBS does not appear to have given any significant change in magnitude of cure retardation. All results agreed very well with the cure behavior as discussed previously in Figure 8.

Mechanical properties of EPM vulcanizate

Table II depicts the mechanical properties of the EPM vulcanizates modified with various types and loadings of interaction modifiers. It is evident that all mechanical properties, especially tensile strength and elongation at break, increased significantly with increasing *p*-PDA content. Since the addition of *p*-PDA up to 0.16 phr caused no significant change in crosslink density (as evidenced from the cure curve characteristics), it could be summarized that the enhancement in mechanical properties found in the system with *p*-PDA was mainly the result of improvements in rubber–filler interaction and degree of filler dispersion rather than the increased crosslink density. However, the greatest improvement in the mechanical properties found at 0.2 phr of *p*-PDA may have arose from the combined effects of the enhanced filler dispersion, the improved rubber–filler interaction and the increased crosslink density, particularly the co-crosslink between Ricon130MA8 and EPM phases. Unlike *p*-PDA, the presence of TBBS gave a significant improvement in elongation at break in association with a slight enhancement in tensile strength. As TBBS had no significant effect on crosslink density, the improvement in tensile

strength might be attributed to the improved filler dispersion and the enhancement of rubber–filler interaction. Surprisingly, the modulus of the vulcanizate was found to reduce with increasing TBBS content. The reduction of modulus is thought to be the consequence of improved filler dispersion which dominates the small improvement of rubber–filler interaction found when TBBS is used as a modifier.

CONCLUSION

The attempt to develop interaction between ethylene-propylene rubber (EPM) and carbon black (CB) as reinforcing filler was conducted by the use of either *p*-PDA or TBBS as interaction modifiers.

Results of the CB-M/B suggest that BRC as an indication of rubber–CB interaction was increased with increasing loading of the interaction modifiers, especially *p*-PDA. Magnitude of the Payne effect decreased with the increased loading of *p*-PDA which was in line with the BRC data. The enhancements in mechanical properties particularly tensile strength and elongation at break of EPM vulcanizates modified with *p*-PDA were observed. Compared with *p*-PDA, TBBS has a lesser effect on the mechanical properties of the vulcanizates. Such enhancements were mainly governed by the improved interaction between CB and the EPM matrix.

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