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Mechanical Properties of Industrial Tyre Rubber Compounds

Mohd Bijarimi, H. Zulkafli and M.D.H. Beg
Universiti Malaysia Pahang, Lebuhraya Tun Razak, 26300 Gambang, Kuantan, Pahang, Malaysia

Abstract: This study aims to investigate the effect of carbon black structures on the mechanical properties of industrial tyre rubber compounds containing Natural Rubber (NR) and Styrene Butadiene Rubber (SBR). Different carbon black structures were used and characterised with respect to their rheological and physical properties. It was found that the NR compound containing high structure black, i.e., N550 showed higher torque and shorter cure times. In contrast, the NR/SBR blends compound did not clearly influence this phenomenon. It was also found that the carbon black grades of N375 and N339; despite of different carbon black structures showed a little influence on the physical properties such as hardness, rebound resilience, tensile and tear strength in NR/SBR blends compound.

Key words: Carbon black, hardness, tensile, tear, resilience

INTRODUCTION

Rubber being one of the most outstanding materials is widely used in many engineering applications such as automotive, civil and electrical. It has been well established that rubber without filler materials have very low physical strength and of no practical use (Edwards, 1990; Kamal *et al*., 2009). Fillers are compounding ingredients added to a rubber compound for the purpose of either reinforcing or cheapening the compound. Despite that, fillers can also be used to modify the physical properties of both unvulcanized and vulcanized rubbers. Typical filler materials include carbon black, calcium silicate, calcium carbonate and clay. Fillers can be classified as black or white (non-black) fillers. Black fillers are more widely used in the rubber industry than white fillers. It is used in tyres, hoses and cable industries. Meanwhile, white fillers are used in the footwear, general rubber goods and automotive industries. Apart from the traditional white fillers, there appears to be a lot of works on the use of biomass materials such as oil palm waste, rice husk (Sae-Oui *et al*., 2002; Sarkawi and Yusuf, 2003; Arayapranee *et al*., 2005) as fillers in the rubber compound lately.

Carbon black is the most popular filler added into the rubber compound due to its ability to enhance the strength properties of rubber vulcanizate as compared to gum vulcanizate; where no filler added (Baker, 1978; Bagghi and Sharma, 1981; Edwards, 1990). Generally, there are various types of carbon black grades used in the rubber industry such as N-200 ISAF (Intermediate Super Abrasion Furnace), N-300 HAF (High Abrasion Furnace), N-500 FEF (Fast Extruding Furnace) and N-660 GPF (General Purpose Furnace) series. However, the choice of carbon black grades for any given rubber formulation must take into account the desired physical properties of the end products, processing methods and costs.

Numerous studies have been found on the addition of carbon black in the rubber compounds as its ability to enhance the mechanical properties (Medalia, 1974, 1978; Medalia and Avrom, 1987; Kraus, 1984; Gessler *et al*., 1978). However, there were still not much works on the effect of carbon black grades in the tyre tread compound apart from the limited number of works (Patel and Byers, 1980; Wang *et al*., 2001; Chung *et al*., 2002). As far as the authors are aware, so far there is no specific works done to investigate the how the roles of carbon black structures influence the uncured and cured compounds. This study gives an insight on how different structures of carbon black influenced the rheological and physical properties of the rubber vulcanizates in both NR and NR/SBR blends compound.

MATERIALS AND METHODS

Materials: In this study, Natural Rubber (NR) and NR/styrene Butadiene Rubber (SBR) blend was used to investigate the effect of cure characteristics and mechanical properties of rubber compounds. The carbon black grades used were N339/N375 and N550/N660 supplied by Cabot Corporation, Malaysia and the study was conducted in 2009.

Table 1 shows the typical industrial tyre tread compounds, formulations A1, B1 and C1 are standard

Corresponding Author: Mohd Bijarmi Mat Piah, Faculty of Chemical and Natural Resources Engineering, Malaysia
Tel: +609-5492918 Fax: +609-5492889

recipes meanwhile recipes A2, B2 and C2 are variants. The effects of carbon black N550/N660 and N339/N375 were investigated in the formulations A1/A2 and B1/B2 and C1/C2, respectively.

Methods: Compounds were prepared in a two-step process; preparation of the masterbatch in a 1.6L Banbury internal mixer (BR1600) and addition of curatives on a two-roll mill.

As shown in the Table 2, the rubber was broken down in the internal mixer for 50 sec at 60 rpm. Half of the masterbatch ingredients were later added and mixed for another 50 sec. Subsequently, the carbon black was added and followed by the remaining ingredients. Finally the compound was dumped and weighed. The compound was cooled to room temperature for 3 h before mixing with sulphur and accelerator. The mixing was carried out on a two-roll mill with roll speed of 30 rpm. After the addition of sulphur and accelerator, the rubber band was cut and folded several times to ensure uniform and homogeneous mixing. Total milling time was about 10 min. The compound was stored at room temperature for 24 h before moulding.

The rheological properties of all compounds were determined by using an oscillating disc Monsanto Rheometer MDR 2000 and Mosanto Viscometer VM2000 according to the ISO 289 and ISO 3417, respectively. The rheological such as cure characteristics and physical properties of the rubber vulcanizates were measured.

Table 1: Compound recipes for all formulations

	A1	A2	B1	B2	C1	C2
Ingredients	(pphr)					
NR	100.0	100.0	80.0	80.0	72.0	72.0
SBR	-	-	20.0	20.0	28.0	28.0
N375	-	-	57.0	-	55.0	-
N339	-	-	-	57.0	-	55.0
N550	53.0	-	-	-	-	-
N660	-	53.0	-	-	-	-
Zinc oxide	5.0	5.0	4.5	4.5	3.0	3.0
Stearic acid	2.0	2.0	2.0	2.0	4.0	4.0
Antioxidant	2.0	2.0	1.0	1.0	0.5	0.5
Antiozonant	1.0	1.0	2.0	2.0	2.0	2.0
Aromatic oil	-	-	-	-	10.0	10.0
Sulphur	2.5	2.5	2.0	2.0	1.9	1.9
Accelerator	1.5	1.5	0.8	0.8	0.8	0.8
Retarder	0.4	0.4	0.2	0.2	0.4	0.4
Total	167.4	167.4	169.5	169.5	177.6	177.6

Table 2: Compounding mixing cycles for all rubber compounds

Mixing steps	Ram down time (sec)	Step instructions
1	0	1st addition: Rubber
2	50	2nd addition: Chemicals
3	100	3rd addition: Carbon black
4	150	4th addition: Chemicals
5	210	Brush/sweep
6	230	Discharge

Cure characteristics at 150°C

- Minimum torque (ML) (dNm)
- Maximum torque (MH) (dNm)
- Optimum cure time, TC (95) (min)
- Mooney Scorch at 130°C
  - Scorch time at (t5) (min)
  - Scorch time (t35) (min)

For the evaluation of the physical properties, all compounds were cured to their respective optimum cure time TC (95) at 150°C.

Physical properties

- Hardness (Shore A)
- Rebound resilience
- Tensile strength (MPa)
- Tear strength (N mm^{-1})

RESULTS AND DISCUSSION

Rheological properties: The rheological properties i.e., minimum torque (ML) and maximum torque (MH) tested at 150°C, mooney viscosity ML1+4@100°C and scorch @130°C are shown in Table 3.

The maximum torque for all the compounds tested in this study varies from 12.95 to 23.26 dNm. In compound A2 where a low structure carbon black filler, i.e., N660 was used, the maximum torque decreased as compared to A1 i.e., from 23.26 to 20.59 dNm. This could be attributed due to higher structure of N550 in the formulation A1 which contains only NR. Nevertheless, this is not the case for NR/SBR blends i.e., compounds B and C where the MH values are very close to each other as shown in Table 3. Ciesielski (1990) reported that the torque difference is an indirect indicator of crosslink density of rubber composites. It can be said that the higher torque in compound A1 is related to the relatively higher crosslink density and thus the hardness.

As shown in the Table 3, the cure time TC (90) of the compounds containing lower structure black, i.e., A2, B1

Table 3: Compound rheological properties, viscosity and scorch time

Ingredients	A1	A2	B1	B2	C1	C2
ML (dNm)	2.23	1.92	3.43	3.91	2.46	2.27
MH (dNm)	23.26	20.59	20.12	20.99	13.53	12.95
TC(10) (min)	6.48	4.77	4.73	5.27	8.38	6.85
TC(40) (min)	7.63	5.77	6.88	7.77	12.28	10.08
TC(90) (min)	12.02	9.37	14.82	17.37	22.87	18.55
TC(95) (min)	14.14	11.08	18.28	22.00	27.02	21.58
ML 1+4 (100°C)						
Scorch @ (130°C)	64.3	55.1	81.3	92.1	54.5	51.2
t5 (min)	12:16	12:23	12:51	14:20	23:11	21:22
t35 (min)	14:18	14:25	14:49	16:38	27:49	25:06

Table 4: Physical properties of compounds cured at 150°C

Ingredients	A1	A2	B1	B2	C1	C2
Hardness (Shore A)	69	66	68	71	61	59
Rebound resilience	51.2	57	38.9	38.4	34.2	34.5
Density (g cm^{-3})	1.145	1.14	1.149	1.154	1.13	1.126
Tensile strength (MPa)	16.5	16.6	20.9	21.2	18.5	18.7
Elongation at break (%)	289	345	401	385	546	576
S200 (MPa)	12.4	9.8	9.8	10.9	5.2	4.5
S300 (MPa)	-	15.8	16.8	18	9.6	8.6
Tear strength (N mm^{-1})	15.9	12.8	24.2	25	54.2	58.5
Heat build-up (°C)	16	13	26	27	27	27
Heat build -up set (%)	2	1.5	4.5	5	7.5	7

and C1 are shorter as compared to those of higher structure black in compounds A1, B2 and C2. It was also observed that the scorch times were not much affected by the carbon black grades tested in this experiment.

Physical properties: It has been well established that the incorporation of carbon black into rubber compound generally improve the strength, extensibility, fatigue resistance and abrasion resistance. In this experiment we are concerned with hardness, rebound resilience, tensile strength and tear strength only. The physical properties such as hardness, rebound resilience, tensile strength and tear strength of all compounds are shown in the Table 4.

Hardness: Hardness is a measure of the resistance to a reversible deformation of the rubber by a rigid indentor and widely used as a quality control measure. The hardness was measured using a Wallace Shore A durometer according to ISO 48. In this experiment, the results are shown in Table 4. The hardness values vary from 59 to 71 units for all compounds.

In compound A2, the hardness value was lower by 3 units as compared to A1 which is due to the substitution of carbon black N550 to N660. This softness could be due to relatively lower carbon black structure of N660 as compared to the N550. Structure describes the number of particles that fuse to form an aggregate. If the primary aggregate is composed of many prime particles with considerable branching and chaining, it is referred to as a high structure black, where the structure is expressed in terms of Dibutyl phthalate absorption value (Mahapatra *et al*., 2004). The more particles in the aggregate, the more complex in the shape and greater void volume created. These voids can be filled with polymer (Laube *et al*., 2001). In contrast, the substitution of N375 to N339 resulted an increase of hardness value by 3 units. Similarly, this can also be explained that the N339 has higher structure as compared to N375. However, the opposite phenomenon was found to be occurred in the compounds C1/C2 where the hardness decreased by 3 units even though the relatively higher structure of black, i.e., N339 was used. This can be explained by the addition of oil in the compound C1/C2 probably has influenced the hardness value. In the absence oil, it was observed that the use of relatively higher structure of carbon black has contributed to a certain extent of higher hardness value. In addition, the maximum torque is correlated with hardness and modulus (Teh *et al*., 2003). From Table 3, the maximum torque was higher in A1 and hence the increase of hardness was expected.

Rebound resilience: The rebound resilience test was carried out in accordance the ISO 4662:1986 test method. Rebound resilience is the ratio of the energy of the indentor after impact to its energy before impact expressed as a percentage. Generally, the rebound resilience values did not show significant effect on different carbon black grades tested in this experiment as shown in the Table 4. However, this could only be true in the B1/B2 and C1/C2 compounds. In compound A2, where the carbon black grade was substituted from N550 to N660, the resilience increased by about 6 units. Perhaps this could be due to the drop of hardness value hence contributed to the higher value of rebound resilience. Rios *et al*. (2001) found that there was an inverse relationship between hardness and resilience. If the hardness achieved was high, the resilience decreased because there were more points of slippage between the reinforcement and the matrix and also because of the reinforcing material tends to clump together so that particles touch one another instead of being totally embedded in the rubber matrix.

Tensile strength: According to Edwards (1990) the term reinforcement by the carbon black filler will refer simply to the striking changes in stress-strain properties brought about by the presence of reinforcing particles in the rubber vulcanizate. Table 4 shows the effect of different carbon black grades tested against the tensile properties in the tread compounds measured according to ISO 37:1994. Surprisingly, the tensile strength values showed almost no effect due to the substitution of N375/N339 and N550/N660 carbon black in all the formulations.

This finding is consistent with Baker (1978) where they found that the two improved blacks, N339 and N375, showed no distinct advantages over the two normal blacks, N220 and N330, in the vulcanizate properties examined. The N339 black deviates slightly from the general trend shown by the others in tensile strength and elongation at break, showing slightly lower values than the other blacks at lower loadings, but the differences are not large.

Tear strength: The tensile strength values for all compounds vary from 16.5 to 21.2 MPa as shown in Table 4. There were very minimal effect to be observed to the compounds A1/A2 and B1/B2. However, in compound C2 the tear strength value increased by 4 units. Unlike the tensile strength values, the compounds C1/C2 appeared to having very much higher tear strength than A1/A2 and B1/B2. This can be explained by the relatively lower hardness values in compounds C1/C2 against compounds B1/B2. Furthermore according to researchers, a decrease in particle size results in increase in tear strength. This was apparent in the compound C2 where carbon black of N339 was used instead of N375. However, the same phenomenon did not happen in the compound B2 despite the same N339 grade was used.

CONCLUSIONS

From the observations and results obtained, the following conclusions can be drawn:

- The NR compound containing high structure black i.e., N550 showed higher torque and shorter cure times as compared to low structure black of N660 grade. However, this effect was not observed in the NR/SBR blends compound tested in this experimental works
- The use of low and high structures black grades i.e., N375 and N339 showed a little influence on the physical properties such as hardness, rebound resilience, tensile and tear strength in NR/SBR blends compound.

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