The influence of natural rubber – butadiene rubber and carbon black type on the mechanical properties of tread compound

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Abstract. The objective of this study is to optimize the performance of the tread compound, by mixing of specified ingredients, such as Natural Rubber (NR) and Butadiene Rubber (BR) at various compositions, and also the type of carbon black as filler. The composition of NR and BR are used as follows 100:0 ; 95:5 and 85:15. Other materials used as rubber additives are peptizer, ZnO, stearic acid, additives, processing oil, and curatives. Two types of carbon black namely N220 and N234 are used in this research. The un-vulcanized rubber from each formula was characterized of curing characteristics and Mooney viscosity. The vulcanized rubber was then characterized its mechanical and thermal properties. It is clear from the research that, the addition of BR into the NR compound increases the abrasion resistance, rebound resilience and hardness, yet the glass transition temperature of the rubber compounds decreases. On the other hand, it seems that the rubber compound reinforced N220 or N234 gives comparable results on both mechanical and thermal properties.

1. Introduction

Regarding its vast improvement in the last decades, tire industry has always won the first place considering the volume of the studies done on the properties of rubber. Many researchers, from all over the world, are trying to present new products with higher capabilities and efficiency.

From among these studies, the most has been dedicated to the tread of the tire for this is the part of the tire which has the greatest impact on the way and the type of use. The performance of tire tread was measured by combination of “magic triangle properties”. They are rolling resistance, traction and wear. The balance of those properties increases the quality of tire tread [1].

Shiva and Atashi (2010) [2] have studied the optimization of failure properties of the Passenger Tire Tread Compound made of a compound of SBR with NR and BR, by altering curing condition and modeling rubber's behavior. Pham et al. studied the blends of two types of SBR with NR and the inherent properties of rubbers on the properties of the final compound [3]. The results showed an increase in Mooney viscosity and scorch time as the SBR increased in the compound. Kaushik et al., (2010) [4] studied the compound of NR/BR/HSR with the presence of different amounts of carbon black and found that the samples gained a very good abrasion resistance and in addition, as the size of carbon black particles decreased and became close to nano particle size, the strength of the samples increased.
Since tire is a composite structure made of substances with various properties, choosing each of these substances and the amount of each of them requires a rich knowledge. One of the substances which are added to the rubber compounds are reinforced fillers and have an important effects on the properties of the final product. Carbon black is one of the reinforcements that frequently used in tire industry and many researchers have studied its effect on mechanical and dynamic properties of tires. The size of carbon black particles and its specific surface are the most important factors affecting on tires’ eventual properties [5].

Besides of reinforcing filler, rubber part has main role to the performance of tire tread. Cured BR imparts excellent abrasion resistance (good tread wear), and low rolling resistance (good fuel economy) due to its low glass transition temperature (Tg). In this study, optimization of BR composition in NR compounding formulation was studied to increase the abrasion resistance and tire retread performance. Instead of that, type of carbon black was also observed to proof the hypothesis that nano particle size carbon black would increase the strength of rubber compounding. Detail research on properties of compounding were performed without silica.

2. Experimental

All mixing ingredients were used as received. Natural rubber (NR) as Ribbed Smoked Sheet (RSS1) type with the density of 0.95 was provided by local supplier in Indonesia. Butadiene rubber (BR) with the Mooney viscosity 40 and the density of 0.92 was obtained from Goodyear Chemical. Carbon black N 220 and N234 were purchased from Cabot.

2.1 Preparation and Testing of Rubber Compound

A laboratory-sized kneader Moriyama DS3-10MWB-E was employed to prepare rubber compounds. The mixing was divided into two stages. In the first stage, the mixing condition was set at the temperature of 100°C and the velocity of 60 rpm. The mixing was started by mixing NR or NR-BR at various compositions along with plasticizers for about 5 min, and followed by adding zinc oxide, stearic acid, carbon black, other additives and a half of oil for about 6 min. The remaining oil was then added into the kneader for about 2 min. The compound was passed on the two roll mill at 70°C for 6 times. After the master batch was stored for a night to allow the rubber compound to rest, the batch then was mixed with curatives in the kneader. The temperature was set at 70 °C and the velocity of 60 rpm for about 2 min. At the end of the mixing, the batch again was passed on the two-roll mill, cooled and stored for another 24 h before it can be used for further analysis.
Six formulations were designed according to Table 1. In all formulations the total of rubber (NR and BR) was kept at 100 phr. Cure characteristics were determined using an MDR 3000 MonTech. Delta torque is obtained by subtracting of maximum torque (MH) and minimum torque (ML). Scorch time ($t_{s2}$) is the time to reach 2 units above minimum torque (ML) and optimum cure time ($t_{90}$) is the time to achieve 90% of delta torque above minimum. Sheets and test specimens were vulcanized by a compression molding Dr. Collin. The experimental curing press was set at the temperature of 140ºC and the time of curing was based to $t_{90}$ as shown in Table 2, which were obtained from curing testing. Mooney viscosity was measured using a MV 2000 MonTech at 100ºC, one min of preheating and four minutes of rotation ML (1+4)100 ºC.

| Component | FR1 | F1 | F2 | FR2 | F3 | F4 |
|-----------|-----|----|----|-----|----|----|
| NR        | 100 | 95 | 85 | 100 | 95 | 85 |
| BR        |     | 5  | 15 |     | 5  | 15 |
| Peptizer  | 3   | 3  | 3  | 3   | 3  | 3  |
| ZnO       | 5   | 5  | 5  | 5   | 5  | 5  |
| Stearic acid | 3 | 3  | 3  | 3   | 3  | 3  |
| N220      | 50  | 50 | 50 |     | 50 | 50 |
| N234      |     | 50 | 50 |     | 50 | 50 |
| Additives | 8   | 8  | 8  | 8   | 8  | 8  |
| Processing oil | 6 | 6  | 6  | 6   | 6  | 6  |
| Curatives | 3   | 3  | 3  | 3   | 3  | 3  |

| Compound name | ML (dNm) | MH (dNm) | Delta Torque (MH – ML) | $t_{s2}$ (min) | $t_{90}$ (min) |
|---------------|----------|----------|------------------------|----------------|----------------|
| FR1           | 2.65     | 15.66    | 13.01                  | 3.23           | 9.47           |
| F1            | 2.43     | 14.55    | 12.12                  | 3.62           | 8.84           |
| F2            | 2.70     | 15.25    | 12.55                  | 3.58           | 8.65           |
| FR2           | 2.48     | 14.38    | 11.91                  | 3.87           | 9.10           |
| F3            | 2.44     | 13.56    | 11.12                  | 3.08           | 7.88           |
| F4            | 2.47     | 14.99    | 12.52                  | 3.81           | 8.74           |

2.2 Testing of Rubber Vulcanizates

Tensile strength was determined according to ASTM D412 by using Go Tech 7049 Universal Testing Machine 2kN, Taiwan. Abrasion resistance was performed with DIN abrasion tester machine AT150 as per DIN ISO 4649. Rebound resilience and hardness according to ASTM D2632 and ASTM D2240, respectively. The glass transition temperature (Tg) was measured using a Differential Scanning Calorimetry (DSC) Setaram, France according to ASTM D 3418-15.
3. Results and Discussion

The processing characteristics and physical mechanical properties of rubber compounds have been presented in graph or table below.

3.1 Unvulcanized Rubber Curing Characteristic and Mooney Viscosity

It can be seen from Figure 1 and Table 2 that the addition of BR into the NR compound does not have a significant effect on altering curing time, scorch time or delta torque. There is no significant difference of curing characteristics between rubber compound containing N220 and N234.

![Figure 1. The optimum cure time and the scorch time of rubber compounds](image1.png)

From Figure 2, the addition of BR into the rubber compound slightly lowering the Mooney viscosity (MV). It is because the MV of BR is lower than NR due to low molecular weight of BR compared to NR [6]. There is no significant difference of Mooney Viscosity between rubber compound reinforced N220 and rubber compound reinforced N234 since both types of carbon black has similar particle size and gave the same effect to the viscosity of compounding.

![Figure 2. Mooney viscosity of rubber compounds](image2.png)
3.2 Characterization of Vulcanized Rubber

Hardness

Hardness may be defined as a material’s resistance to permanent indentation. The Shore A Hardness Scale measures the hardness of flexible mold rubbers that range in hardness from very soft and flexible, to medium and somewhat flexible, to hard with almost no flexibility at all. It is expected that the addition of BR will not have a significant effect on altering the hardness of rubber compound. Yet in this research, the addition of BR decreased the hardness of rubber compound as presented in Figure 3. Hardness of rubber compound containing N220 is slightly higher than rubber compound containing N234 since lower particle size of carbon black could lead better dispersion level and increase the hardness.

![Figure 3. Hardness of rubber compounds](image)

Tensile Strength

Tensile strength is the maximum tensile stress reached in stretching a test piece (either an O-ring or dumbbell). Tensile tests are used for controlling product quality and for determining the effect of chemical or thermal exposure or an elastomer. In the latter case, it is the retention of these physical properties, rather than the absolute values of the tensile stress, elongation or modulus, that is significant. Figure 4 shows the result of tensile strength measurement. The strength of rubber compound decreases significantly with the addition of 15 phr BR into the rubber compound. Compared to FR1 dan F1, F2 has lower tensile strength. So does F4 that showed significant low tensile strength after addition of BR. It is because unlike NR, BR does not have crystallization properties; therefore, the tensile strength of BR is very poor [7]. A not significant increasing point was observed in addition of N220 filler that gave higher tensile strength than N234.
Abrasion resistance is the ability of a material to resist mechanical action such as rubbing, scraping, or erosion that tends progressively to remove material from its surface. When a product has abrasion resistance, it will resist erosion caused by scraping, rubbing, and other types of mechanical wear. This allows the material to retain its integrity and hold its form. This can be important when the form of a material is critical to its function, as seen when moving parts are carefully machined for maximum efficiency [8]. Abrasion resistant materials can be used for both moving and fixed parts in settings where wearing could become an issue.

The properties of a vulcanized rubber can be significantly influenced by details of the compounding. Practical materials will have, in addition to the base polymer, fillers, antioxidants, crosslinking agents, accelerators etc. All of these can have an influence on the physical and chemical stability of the finished material. For example, rubber abrasion resistance can be related quantitatively to the frictional force and the crack growth characteristic of the rubber [9].
Abrasion behavior was presented by ARI% as shown in Figure 5. Along with the decreasing of hardness, BR substitution leads to higher abrasion resistance. Both rubber compounds containing N220 and N134 give a comparable value of abrasion resistance.

**Rebound Resilience**

Resilience of a rubber compound is a measurement of how elastic it is when exposed to various stresses. Resilience is the ratio of energy released in deformation recovery to the energy that caused the deformation. Measurement of rubber resilience can assist in choosing the right material for a given application, for example tread tyre.

The increasing of BR addition leads to resilience percentage improvement. It was in agreement with the decreasing of hardness and increasing of abrasion resistance [10]. Addition of N220 and N234 did not give any significant difference to the result of resilience testing.

![Rebound Resilience of Rubber Compounds](image)

**Figure 6.** Rebound resilience of rubber compounds

**The Glass Transition Temperature**

Glass Transition is a method to characterize a property of a polymeric material. The glass transition is the temperature where the polymer goes from a hard, glass like state to a rubber like state [11]. Due to the result of mechanical properties above, the measurement of Tg were performed to F1, F2, F3, and F4 samples, which is consist of blended NR and BR. The result is shown in Table 3.

| Compounding | Tg (°C)    |
|-------------|------------|
| F1          | -58.89     |
| F2          | -59.44     |
| F3          | -58.95     |
| F4          | -59.53     |
Cured BR imparts excellent abrasion resistance (good tread wear), and low rolling resistance (good fuel economy) due to its low glass transition temperature (Tg). The low Tg, typically <–90°C, is a result of the low “vinyl” content of BR. However, low Tg also leads to poor wet traction properties, so BR is usually blended with other elastomers like NR for tread compounds. It is expected that the addition of BR will have a significant effect on lowering the Tg of rubber compound [12]. Yet in this research, the addition of BR up to 15 phr only lowers the Tg by 1 degree compared to rubber compound containing 5 phr of BR. There is no significant difference on the Tg between rubber compound containing N220 and rubber compound containing N234 [13].

4. Conclusion
A blend of natural rubber (NR)/cis-butadiene rubber (BR) in the presence of carbon black filler showed excellent mechanical properties of a tread formulation. The mechanical properties such as abrasion resistance and rebound resilience have been improved with the presence of cis-butadiene rubber. Here the presence of carbon black N220 could give slightly better mechanical properties compared to carbon black N234 due to the better dispersion of N220 into the rubber compound. Due to good properties of these blending, it is possible to apply this formulation in tread tyre. Detail testing in order to fulfill the requirement of tread compound should be performed in advanced.

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