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Effect of Brown Crepe Rubber Content on Cure Characteristics and Physical Properties of Tire Retreading Compound

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ABSTRACT

Natural Rubber (NR) based products manufacturing is one of the main industries in Sri Lanka. Due to high demand of petroleum-based synthetic rubbers, the price and demand for NR have also augmented. Therefore, local rubber industries practice replacement of Ribbed Smoked Sheets (RSS) grade rubber partly with brown crepe (BC) grade of NR in their rubber compounds. Since BC rubber is contaminated with foreign particles such as dust, soil and sand, it is regarded as a low-in-quality cheaper grade. It is reported that certain local rubber industries do not concentrate on the quality aspects of rubber products in adding BC into compounds, since their primary aim is to attract higher profits. This is a problematic situation, today. Since retreading of worn-out tires is a leading sector of the local rubber industry, the local rubber landscape is considerably affected with this problematic development. In this study, the tire retreading compound s based on 70/30, 80/20 and 90/10 ratios of RSS / BC rubber were investigated to identify the most suitable and economical amalgamation. BC content could not be increased more than 70/30, due to non-uniform blending of two grades. Cure characteristics, tensile strength, percent elongation at break, modulus at 300% elongation and physical properties such as hardness, resilience, compression set, age resistance, abrasion resistance and flex cracking resistance were tested for all these compounds with the control compound benchmarked with RSS grade. The best properties were shown for the control compound, and all test properties were gradually reduced with increase of BC percentage. Most of the test properties of the compound with RSS/BC ratio at 70/30 were unacceptable while other compounds were at acceptable levels. Thus, RSS/BC at 80/20 is the optimum ratio for formulating tire retreading compounds with a suitable balance of cost and technical properties.

KEYWORDS: *optimum percentage, ribbed smoked sheets, brown crepe rubber, technical properties, tire retreading compounds*

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1 INTRODUCTION

Natural rubber has been known for many centuries and it is obtained from latex, collected in more than thousand species of trees. Among all species, *Heavea brasiliensis* is the main source of commercial rubber today, due to its high productivity and economics (Alahapperuma 2007, Rubber Research Institute 2003, Venei 2007). The plant *Heavea brasiliensis* is indigenous to Brazil and South America. In the contemporary, Thailand, Indonesia, Vietnam, China, India, Malaysia and Sri Lanka have been the major sources of NR, and total world production is closer to thirteen million metric tons (Statista 2021).

In Sri Lanka, NR is produced generally in form of blocks and sheets. Grades available in sheet form are RSS, BC (sometimes called as scrap rubber), sole crepe and pale crepe etc.. Production of rubber in block form differs technically from sheet form. Of these grades, block rubber, sole crepe and pale crepe grades are especially produced for the export market. The remaining two, i.e. RSS and BC grades, are mainly consumed by local industries. Of the two grades, RSS grade is produced from coagulum of latex, after smoke drying under control processes, whereas BC rubber is prepared by mixing pre-coagulated dried cup lumps, tree laces, shell scrap and earth scrap collected from plantation sites. When earth scrap is used, it is washed and dried before further processing. However, BC rubber is contaminated with bark, dirt, soil and sand particles, and is regarded as a cheap and low-quality grade (Alahapperuma 2007, Eastern Merchants PLC 2018, Subramanium 2002, Tillekeratne, Nugawela & Seneviratne 2003). Therefore, RSS is the preferable grade for producing good quality rubber products in the local industries (Alahapperuma 2007, Rubber Research Institute 2014).

Grading of both types, RSS and BC, is based on visual inspection and dirt content. RSS is the highest in demand grade of NR, in Sri Lanka. There are five different categories of RSS namely, RSS No. 1 to RSS No. 5. The grade of RSS No.1 is the apical, and rubber quality decreases as grade number increases (Rubber Research Institute 2014). BC produced in Sri Lanka consists of higher amounts of bark and volatile matter, and lacks consistency in quality. However, quality of BC mainly depends on method of manufacture and type of scrap used. After processing, BC is categorized into five grades namely, 1X (FAB+), 2X (FAB), 3X (FAB-), Scrap Crepe No. 4 and Standard Flat Bark Crepe (FB). The grade 1X (FAB+) is of the best quality and for the remainder, quality is in descending order (Tillekeratne, Nugawela & Seneviratne 2003, Rubber Research Institute 2003). Since no sufficient chemical treatment is taking place during production of BC rubber (Subramanium 2002), there is a chance for variation of cleanliness and properties of BC based compounds.

NR is used solely as the base rubber in almost all Sri Lankan rubber industries. However, as the price of RSS grade is in the ascendency due to high demand of synthetic rubbers, it is sold at an exorbitant rate. Thus, continuous usage of

RSS grade alone in local rubber industries is no longer economical. Because of this, the industries have begun to replace RSS grade partly with cheaply available BC rubber in their rubber compounds. It is reported that with the sole aim of getting profits, certain rubber industries blend BC rubber with RSS in arbitrary ratios, even at the expense of quality of products. This is indeed a regrettable situation in local rubber industries of today (Alahapperuma 2007).

Since this practice may deteriorate technical properties of rubber products, prediction of an optimum percentage of BC rubber in compounds will help rubber product manufacturers to preserve quality of their products. Previous studies on effects of addition of lower grade BC rubber to RSS grade are rarely found in the literature.

Tire sector is the main manufacturing segment of Sri Lankan natural rubber products. Retreading is a major sub sector of tire sector (Sri Lanka Export Development Board, 2015), and worn out tires are restored to reusable conditions by consolidation of surface of worn out tires with a specially prepared rubber (retreading) compound (Wikipedia 2021). Due to high production capacity of tire products, adverse effects caused by addition of arbitrary amounts of BC rubber in related formulations is a significant issue of the sector (Alahapperuma 2007).

Therefore, aiming to identify optimum combination of RSS and BC rubber in a typical tire retreading formation (Subramanium 2002), blends containing different proportions of these two grades were tested for cure, physical and mechanical properties, in this study.

2 MATERIALS AND METHODS

2.1 Raw Materials

Sheets of RSS (RSS No. 2) and BC rubber (2X (FAB) were collected from Agalawatta rubber cultivation area. Laboratory grade compounding ingredients of zinc oxide, stearic acid, insoluble sulphur, cyclohexyl 2-benzothiazole sulphenamide (CBS), prevulcanisation inhibitor (PVI), high abrasion furnace (HAF) black, aromatic oil, coumarone indene (CI) resin and isopropyl phenylenediamene (IPPD) were collected from Rubber Testing Laboratory of Department of Chemical and Process Engineering of University of Moratuwa.

Compound formulations given in the table 1, based on the blends of RSS and BC grades of natural rubber were considered for preparing tire re-treading compounds. Weight ratios of RSS and BC in the blends were taken as 90:10, 80:20 and 70:30. BC rubber content couldn't be increased beyond 30% due to difficulties of forming a continuous and uniform sheet with the adulterating compound, of which the product was brittle in nature.

These compounds were prepared by means of laboratory internal mixer and open two roll mill. Order of incorporation of compounding ingredients into formulations is given in the table 2. After mixing, each compound was sheeted out separately in open two roll mill, for testing of its processability and vulcanizate properties.

2.2 Preparation of Compound Sheets

Table 01: Compound formulations based on RSS and BC rubber blends

Components	**Control compound formulation**	**Compound formulation I**	**Compound formulation II**	**Compound formulation III**
RSS rubber	100.0	90.0	80.0	70.0
BC rubber	0.0	10.0	20.0	30.0
Zinc oxide	5.0	5.0	5.0	5.0
Stearic acid	1.5	1.5	1.5	1.5
Insoluble sulphur	2.5	2.5	2.5	2.5
CBS	1.0	1.0	1.0	1.0
PVI	0.3	0.3	0.3	0.3
HAF black	50.0	50.0	50.0	50.0
Aromatic oil	6.0	6.0	6.0	6.0
CI resin	4.0	4.0	4.0	4.0
IPPD	1.5	1.5	1.5	1.5

Table 02: Order of incorporation of compounding ingredients into formulations

Order of Incorporation	**Compounding ingredients**	**Time of incorporation, min**	**Mixing machine**
First	Zinc oxide, Stearic acid, IPPD	01	Internal mixer
Second	Half of carbon black, Half of oil	1/2	Internal mixer
Third	Half of carbon black, Half of oil	1/2	Internal mixer
Fourth	CI resin, PVI	01	Internal mixer
Fifth	Sulphur, CBS	01	Open two roll mill

2.3 Testing of Processability; Cure Characteristics

Cure characteristics were determined according to ASTM D 2084-95 and ASTM D 5289-95 standards by means of laboratory oscillation disc rheometer at the temperature of 150oC. Readings of two test specimens were averaged, for each compound.

2.4 Sample Preparation and Testing of Vulcanizates

Test specimens for each test (except tensile test) were prepared by molding into standard dimensions followed by curing at the optimum curing conditions, obtained for each compound. After the molded test pieces were de-flashed and conditioned, they were transferred for testing of vulcanizate properties. In case of tensile test, test specimens were punched out from vulcanized sheets, using a standard cutter. Relevant vulcanized sheet in each case was prepared by curing at optimum conditions, given above.

2.4.1 Testing of Tensile Properties

Property of tensile strength, percent elongation at break, and modulus at 300% elongation were measured according to ASTM D 412 standard, using a tensometer (Instron 3300, USA) under crosshead speed of 500 mm/min. Before testing, thickness of each test piece was measured using a thickness gauge. Six test specimens were tested for each compound and readings were averaged.

2.4.2 Testing of Hardness (IRHD)

Hardness (IRHD) measurements were done using Dead Load hardness tester according to ASTM D 1415-68 standard. Two test specimens were tested for each compound and readings were averaged as the mean.

2.4.3 Testing of Vertical Rebound Resilience

Measurements of vertical rebound resilience were done according to ASTM D 3632 standard, using Lupke pendulum. For each test specimen, three rebound readings were noted successively, and the median value was taken. The average result of three test specimens were reported for each compound.

2.4.4 Testing of Accelerated Age Resistance (Property Considered Hardness)

Test for age resistance was carried out according to ASTM D 573–99 standard. A normal oven was used for ageing test specimens. During ageing test, renewed air was slowly circulated inside the oven while test specimens were heated at constant temperature of 70^0C, for 72 hours. Percentage ratio of aged hardness to unaged hardness of each test specimen was calculated. Readings of three test specimens were averaged for each compound.

2.4.5 Testing of Compression Set

Test was done according to ASTM D 395-69 standard, using compression set (constant strain) apparatus. Thickness of all test specimens were measured initially, and they were compressed between two plates separated by spacer bars, to ensure constant strain of 25%. After test specimens were kept under that strain for 72 hours at room temperature, they were removed from the device and allowed to recover for 30 minutes at the same temperature. Three test specimens were tested for each compound, and readings were averaged.

2.4.6 Testing of Abrasion Resistance

The test was done according to ASTM D 1630-61 standard, using Din abrasion tester. Before taking measurements, each test specimen was weighed to the nearest 01mg and specific gravity of each compound was measured using a density meter. During the test, drum was rotated at 40 rpm, and test specimen holder was travelled 0.32 m/second for a distance of 40 m. After each test specimen was taken out from the tester, it was cleaned off dust and weighed to the nearest 01 mg. Volume loss in each case was measured using weight difference and density of test specimen. Readings of three test specimens were averaged for each compound.

2.4.7 Testing of Crack Resistance

Testing was done according to ASTM D 813 standard, using De-mattia flexing machine. The test was continued with frequent inspections until first minute sign of cracking was detected in each test specimen. The approximate number of cycles at this point was recorded. When inspection of flexed test specimen, the grips were separated to a distance of 65 mm. Readings of three test specimens were averaged to express the result.

3 RESULTS & DISCUSSION

Results of processability test (cure characteristics at 150^0C) are given in table 3. Compound formulated with RSS alone (control compound) shows all cure characteristics in desired levels. As BC percentage increased, cure characteristics of scorch time (Ts_2) and cure time (Tc_{90}) have decreased. Compound III having BC and RSS in 30:70 ratio is rather unsafe for processing, because of short delay time and short scorch time.

Table 03: Processability test results (Cure characteristics at 150^0C) of compounds prepared with RSS/BC rubber

Cure Characteristic	**Control compound**		**Compound I**		**Compound II**		**Compound III**	
	Ave.	**SD**	**Ave.**	**SD**	**Ave.**	**SD**	**Ave.**	**SD**
Scorch time (Ts_2) (min)	2.28	0.028	2.08	0.014	1.88	0.056	1.44	0.042
Cure time (Tc_{90}) (min)	18.35	0.035	17.50	0.042	16.79	0.014	15.52	0.056

Note: Ave. – Average, SD - Standard deviation

Faster curing tendency of compound III could be due to higher amount of non rubber constituents, including fatty acids and nitrogen containing substances. As field latex is chemically treated with a basic substance for preservation, protein and phospholipid layers of latex are hydrolyzed into salts of fatty acids, polypeptides and choline. Therefore, amount of nitrogen and fatty acids contained in BC rubber is essentially higher than high quality rubber like RSS and crepe rubber (Subramanium 2002). Besides, BC contains higher amount of highly contaminated foreign matter, such as sand, soil, dirt and bark shavings, gathered from plantation sites (Alahapperuma 2007, Subramanium 2002).

Different studies (Abd-Ali 2014, Nayanashree & Thippeswamy 2013, Trang et al 2013) have demonstrated that microorganisms such as fungi and bacteria react easily with NR due to organic matter such as carbohydrates, proteins and lipids etc. Abd-Ali (2014) further showed that fungal growth can cause changes in its structure including chain cracking, due to breakage of C-H or C-N bonds. According to another study (Onyeagoro, Ohaeri & Timothy 2012), certain bacteria also degrade NR molecules. Thus, due to insufficient treatment with chemicals, there is a propensity for BC rubber to be attacked by microorganisms causing unfair cure properties.

Contaminated foreign matter in BC rubber have been taken as reason for the collapsing nature of interactions in between rubber molecules themselves, and interactions between rubber molecules and carbon black particles, causing lower cure properties in compounds with higher BC, preferably in compound III.

Results of mechanical and physical properties of vulcanizates are given in the table 4. Compound with RSS alone as the base rubber (control compound) possesses desirable mechanical and physical properties. It has shown desirable tensile properties, hardness, age resistance, resilience, compression set, abrasion resistance and flexing properties. When BC percentage was increased, physical and mechanical properties were reduced.

Even though properties of tensile strength, elongation at break and modulus at 300% elongation and physical properties of hardness, resilience and age resistance aren't much affected with increase of BC rubber content up to 30%, abrasion resistance, compression set and flexing properties are remarkably affected. It is found that, the latter set of properties are below average and unsatisfactory, especially in compound III having 30% BC rubber.

Due to higher percentage of BC contained in compound III, rubbery properties may be affected with non-rubbery materials. Nature of cross links formed within the rubber matrix may be the main factor that affects the technical properties of tire treads. The nature of strong bonds formed in between rubber molecules or rubber and vulcanization agent (Alahapperuma 2007, Mohamed 2011) may lead to collapsing by introduction of non rubbery constituents into matrix, leading to compounds with poor mechanical properties (Alahapperuma 2007).

Certain study-based predictions are found in literature on effects of non rubber constituents including proteins, lipids and carbohydrates on properties of NR based compounds. According to Bottier (2019), even though non-rubber percentage of NR is low and is around 5%, the contribution of such contaminants to adverse effects on rubbery properties is significant. This idea implies the possibility to impart severe effects, if higher amounts of non rubbery constituents enter into compounds through low quality grades such as BC.

Table 04: Mechanical and physical properties of compounds prepared with RSS/BC rubber

Physical property	**Control compound**		**Compound I**		**Compound II**		**Compound III**	
	Ave.	**SD**	**Ave.**	**SD**	**Ave.**	**SD**	**Ave.**	**SD**
Tensile strength (N/mm^2)	22.66	0.007	21.95	0.007	21.55	0.008	20.46	0.009
Elongation at break (%)	457.25	0.011	438.20	0.007	437.10	0.103	424.0	0.147
Modulus at 300% elongation	14.17	0.013	13.99	0.042	13.98	0.057	12.34	0.38
Hardness (IRHD)	66	0.00	64	0.00	63	0.57	60	0.57
Age resistance (%)	85.0	0.10	84.4	0.06	81.0	0.10	80.0	0.10
Rebound resilience (%)	66	0.00	64	0.00	63	1.00	60	0.57
Compression set (%)	06.50	0.06	07.20	0.00	8.20	0.06	10.40	0.1
Volumetric abrasion loss (mm^3)	113.3	0.06	133.5	0.06	144.0	0.10	153.0	0.20
Flexibility crack resistance, N	80	1.15	70	0.57	60	1.0	40	1.15

Note: Approximate number of cycles for crack initiation = N* 1000
Ave. = Average, SD = Standard deviation

Another study (Pusca, Bobancu & Duta 2010) revealed that ductile polymer behavior is different to that of brittle polymers, when mineral fillers are incorporated into their mixes. Due to different compositions, same level of ductility cannot be expected in the two grades, RSS and BC. Effects of contaminated sand and other mineral particles in BC rubber might be a reason for undesirable ductility in compound III.

Difficulties experienced in sheeting of compounds having more than 30% BC rubber implies a clear difference in between the two grades.

Usually, attack of microorganisms causes breakage of rubber chains and reduction of mechanical and physical properties. However, Abd-Ali (2014) predicted that any growth of contaminated fungi may cause increase of NR hardness, due to mesh formation by the growing network of fungal mycelia.

Level of reinforcement of carbon black filled rubber compounds is decided based on interaction between carbon black particles and rubber matrix, rubber molecules themselves, and also interaction between carbon black particles themselves, according to the literature (Spahr & Rothon 2016). Nature of interactions between carbon black particles and rubber molecules, and rubber molecules themselves may be disturbed with the adulteration of foreign particles in BC rubber, causing poor strength properties in compounds with higher BC content.

However, according to a recent review article (Robertson & Hardman 2021), real cause for improvement of stiffness of carbon black loaded rubber molecules is not yet proved; it is not universally accepted whether nature of interactions in between carbon black particles and polymer molecules are due to physical adsorption or else, due to chemical bonds that are created during processing of rubber mixes.

Even though organic constituents of NR molecules are regarded as a potential source for degradation of rubber with respect to thermal oxidation of NR mixes, soluble proteins and free fatty acid varieties are a major reason of concern (Tuampoemsab & Sakdapipanich 2007).

Thus, most of the suggestions given in literature imply more chances for non-rubber constituents to hinder favorable cure and vulcanizate properties of rubber compounds that contain larger amount of BC grade natural rubber.

Considering all analyzed compounds, control compound with 100% RSS is the best to formulate rubber retreading mixes. Compound III is rather unsafe with respect to all concerned properties. Due to improving both cost and technical properties, compound II with 20% BC rubber is the most suited economical blend to be used in tire retreading.

4 CONCLUSION & RECOMMENDATIONS

Higher demand for synthetic rubber-based products has resulted in higher prices for NR. Therefore, exorbitant rates have to be paid for quality grades of NR. Of the two main NR grades consumed in Sri Lanka, RSS is the preferable grade to formulate rubber products, considering its better quality parameters. The other grade, BC rubber is considered a lower quality and cheap grade. Higher cost for raw materials has resulted in the search for partial replacement of quality grade of RSS with BC grade.

A substantial proportion of local NR manufactured is taken by tire industry, and tire retreading is an essential sub sector of the local tire industry. Therefore, identifying an optimum proportion of BC rubber in tire retreading compounds is a requirement to prevent undesirable technical properties. Considering cure characteristics, physical and mechanical properties and price levels of two grades, RSS and BC rubber, the ratio of 80:20 is the most economical and desirable ratio to formulate tire retreading compounds.

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