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THE USE OF RECYCLED NATURAL RUBBER POWDER AS FILLER IN NATURAL RUBBER COMPOUNDING.

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Abstract

The use of recycled rubber powder (RRP) in different ratios with carbon black (CB) in compounding natural rubber was investigated in this work. Natural rubber (NR), recycled rubber powder, carbon black and the compounding ingredients were compounded using the two roll mill and the mechanical properties of the blends were analyzed. Six samples A-F were formulated; sample A (0%RRP, 30%CB) sample B (1.5%RRP, 28.5%CB) sample C (4.5%RRP, 25.5%CB), sample D (7.5%RRP, 22.5%CB), sample E (10.5%RRP, 19.5%CB), sample F (13.5%RRP, 16.5%CB). The mechanical properties of most of the vulcanizates decreased with increased recycled rubber powder loading. The hardness values decreased from 19.77 Shore A to 17.63 Shore A from sample A to F, the values of elongation at break from 167.50% to 144.63% from sample A to F and the abrasion resistance from 99.9% to 99.6% from sample A to F. However, the ultimate tensile strength showed improved values i.e 5.70Mpa to 15.30Mpa from sample A to E, with higher rubber powder loading. The fatigue and abrasion resistances increased with increasing RRP loading. It was generally observed that recycled rubber powder holds strong potential in rubber compounding technology as an additive.

1.0 Introduction

The enormous amount of waste tires in the environment has become a major source of concern globally. Tire recycling is capable of generating very large amount of raw materials which can reduce cost as well as environmental pollution. A worn tire, apart from being a secondary source of raw material, also contains a number of chemicals with huge energy potential. Tires contain flexible rubber material whose construction is reinforced with textile and metal materials with natural rubber, soot, steel reinforcements, oils, vulcanizing agents and synthetic rubber [1,2]. When recycling tires, it is necessary to take some measures; reduce the risk of contamination during temporary storage of used tires, increase the usability of used tires, reduce the consumption of raw materials, especially those originating from non-renewable resources, and utilize used tyres as secondary raw materials. [1,3]. Crumb rubber, steel wires, nylon and textile fibres are products resulting from the use of waste rubbers as a raw material source. [5] These materials can be separated from crushed rubber using modern technologies, for the purpose of recycling and further utilization [1,2]. The utilization of the materials from waste tyres that are used repeatedly has been developed for many years, including the applications on road surfaces, the foundations of children playgrounds, sports playgrounds and noise barriers. The use of waste tyres as a substitute for filler in concrete was investigated by Ganjian *et al*. [4] .They used ground rubber powder as a substitute for coarse aggregate of concrete components with 5%,7.2% and 10% composition. Gibala *et al*.[7] reported the rheological properties of composites containing crumb rubber. In his paper, the viscosity of cryogenic powdered composites was lower than that of ambient powdered composites because the shape of the ambient grinding powder was sponge-like and cryogenically ground powder was smooth, hence, it is expected that ground rubber powder will increase the tensile strength.

Surface modification of the crumb rubber has been known to be a very effective method for improving bonding force. Carolina *et al*.[8] investigated effect of chlorination of vulcanized elastomer surface and argued that the most effective result was obtained by chlorination using trichloroisocyanuric acid. Pittolo [9] used a similar surface modification of crumb rubber by chlorination using the same reagent and also by grafting with styrene to enhance the bonding force of the composites. Kim *et al*.[11] compared the mechanical performance and rheological properties of two-phase elastomer blends using various filling techniques. Although much work has been done in this area, problems still remain because of the difficulty in understanding the related surface and inter particle phenomena. Bandyopadhyaey *et al* [13] reported the use of ground crumb rubber in natural rubber base tyre tread cap compound. The results revealed a marginal deterioration in tensile strength as well as a significant decrease in fatigue resistance and abrasion properties of the compound containing crumb rubber. Xipo Zhan *et al* [14], reported the use of reclaimed rubber in Natural rubber Vulcanizates. The cure characteristics of the vulcanizates showed a reduction in the optimum cure time and scorch time. In this work, the use of recycled powdered rubber as a filler was studied. Powdered rubber is unique filler because of its large particle size and low modulus compared to other commercial fillers used in the rubber industry, such as carbon black. [6] . Rubber powder finds application as asphalt modifier, toughening agent for thermoplastics and thermosets or extender/filler in rubber compounds. [10] The main purpose of this work is to use crumb rubber in variable ratios to ascertain its effect on the tensile and mechanical properties of NR vulcanizates.

2.0 Materials and Method

The materials used for this work include Natural rubber (NR) from Rubber Research Institute of Nigeria, Benin, carbon black (CB), zinc oxide, sulphur, stearic acid, trimethylquinoline (TMQ), dithiobisbenzothiazole (MBTS), and recycled rubber powder (RRP) from Lagos, Nigeria. The equipment used include Two roll mill, compression moulding machine, universal tensile machine, Rex durometer, abrasion tester and fatigue tester. The formulation used in this work is presented in the Table 1. All measurements were carried out in parts per hundred of rubber (phr).

Table 1: Compounding formulation.

NR Formulation	A	B	C	D	E	F
%RRP	0	5	15	25	35	45
NR	100	100	100	100	100	100
Carbon black	30	28.5	25.5	22.5	19.5	16.5
RRP	0.0	1.5	4.5	7.5	10.5	13.5
Zinc oxide	4.0	4.0	4.0	4.0	4.0	4.0
Sulphur	2.0	2.0	2.0	2.0	2.0	2.0
Stearic acid	1.5	1.5	1.5	1.5	1.5	1.5
MBTS	1.5	1.5	1.5	1.5	1.5	1.5
TMQ	2.0	2.0	2.0	2.0	2.0	2.0

* RRP (Recycled Rubber Powder)
*MBTS Benzothiazole disulfide (Accelerator)
*TMQ Trimethydihydroquinoline (Antioxidant)

2.1 Filler Characterization

The recycled rubber powder was characterized for its moisture, ash, carbon and nitrogen contents according to Ogunyemi, 2004. [12].

2.2 Compounding process

All materials were weighed. The weighed rubber was masticated using the two roll mill. After proper mastication the compounding ingredients were added in the order of carbon black, stearic acid, zinc oxide and TMQ. Further mastication was carried out for about 35 minutes to allow adequate incorporation of the ingredients until homogeneous product was achieved. The product obtained was then allowed to cool down [14].

2.3 Vulcanization process

This was carried out using the compression moulding machine (model PID 28, Saumya India). The rubber was masticated and vulcanising agents were added in the order of sulphur and MBTS [14]. The heat and pressure molds the rubber to the shape of the cavity and also activates the cure system in the rubber compound. Vulcanization occurred after 3 minutes at a temperature range of 145^{o}C to 155 ^{o}C with a pressure of 1.75 MPa as predetermined from the Rheometer.

2.4 Characterization of vulcanizate properties

The mechanical analysis of the blends were carried out using the universal tensile machine (model UTM192-2L, Saumya India). The sample was fixed to the sample holders, one pulling the sample up and the other pulling it down. As the sample was being stretched by the pulling action of the sample holders, the graphical result containing parameters like yield load, elongation at break (EB), tensile strength at yield load, breaking load and so on was shown on the system. A copy was printed for interpretation and documentation.

2.5 Ultimate tensile strength (UTS)

The ultimate tensile strength was calculated by dividing the maximum load carried by the specimen by the original cross sectional area of the specimen.

2.6 Hardness

This was done using the Rex durometer (model OS-2H, rex guage company, USA). The sample was placed on a metallic base with the indentor pin of the durometer very close to it. The load of the durometer was pressed downward so that the indentor pin could penetrate the sample. The measure of the resistance of the sample to indentation was observed on the display screen and the value was recorded. This was done thrice per sample and the average value was taken.

2.7 Abrasion resistance

The weight of the samples before inserting into the abrasion tester (model ROCKY-DR, Qualitet, India) was measured. The samples were fixed to the sample holder of the abrasion tester and the machine was started, as the sample holder moves, the roller rotates for about 4.2 min for each sample and the final weight of the samples were taken. The final weight was subtracted from the initial weight to get the abrasion resistance value. The following specifications were used:

Abrasion distance of specimen = 40m
Granulation of abrasion sheet = P60
Diameter of specimen = 16mm
Peripheral speed of roller with abrasion sheet = 0.32m/s
Number of revolutions of roller = 40/min
Diameter of roller = 150mm
Lateral speed of specimen per roller rotation = 4.2 min

2.8 Fatigue resistance

This was carried out using a rubber fatigue tester (model ZME-7003, Saumya, India). The sample was clamped tightly to the upper and lower sample holder of the machine and the oscillation is set to 5000. The test process was initiated and the machine started oscillating thereby causing the rubber sample to undergo stress. At the complete oscillation of 5000 per seconds, the machine stopped automatically and the sample was observed for any form of weakness, tear or cut.

3.0 Results and discussion

The results of the characterization of the RRP showed that the % moisture content is 0.54, % ash content is 5.73 while the % carbon content is 52.37 and % nitrogen is 1.26. This results shows that the material has appreciable carbon content for use as filler in natural rubber compounding. The low value of moisture content is of valuable technical advantage during mastication and compounding. The results of the mechanical properties of the composites obtained from the use of RRP as adjunct filler are presented in Table 1. The results in the Table shows that the mechanical properties for majority of the blends except for the ultimate tensile strength decreased with increasing recycled rubber powder loading.

Table 1: Mechanical properties of vulcanizates A to F at different CB and RRP loading.

Samples	A	B	C	D	E	F
Abrasion(%)	99.9	99.9	99.7	99.8	99.8	99.6
Hardness	19.77	16.03	21.40	15.17	19.07	17.63
EB(%)	167.50	141.88	121.38	122.13	259.38	144.63
UTS (MPa)	5.70	5.80	3.50	4.20	15.30	6.50

The decrease in mechanical properties may be due to the relatively larger particle size of RRP usually 100-500 micrometers compared with that of CB and consequently, low surface area. The dilution of reinforcing filler CB with RRP may be responsible for the decrease in the mechanical properties as observed in elongation at break and hardness of the rubber vulcanizates (Figure1) containing them. The vulcanizates A,B,C with minimal or no RRP content showed good mechanical properties, this may be due to the ability of NR to crystallize at high strain [15]. It could be seen that the mechanical properties of sample A with no RRP content for each parameters except ultimate tensile strength showed improved values compared to other blends. This result is also in agreement with the work of Formela and Haponiuk [17] who otherwise suggested that the number of unsaturated bonds should increase with increasing RRP content in the rubber compounds. The unsaturated bonds present in the RRP have ability to form secondary cross- linking thereby competing with the unsaturated bonds in the NR matrix. This fact may support the observed decrease in mechanical properties of the vulcanizates caused by reduced cross- linking density rather than poor adhesion of the rubber matrix.

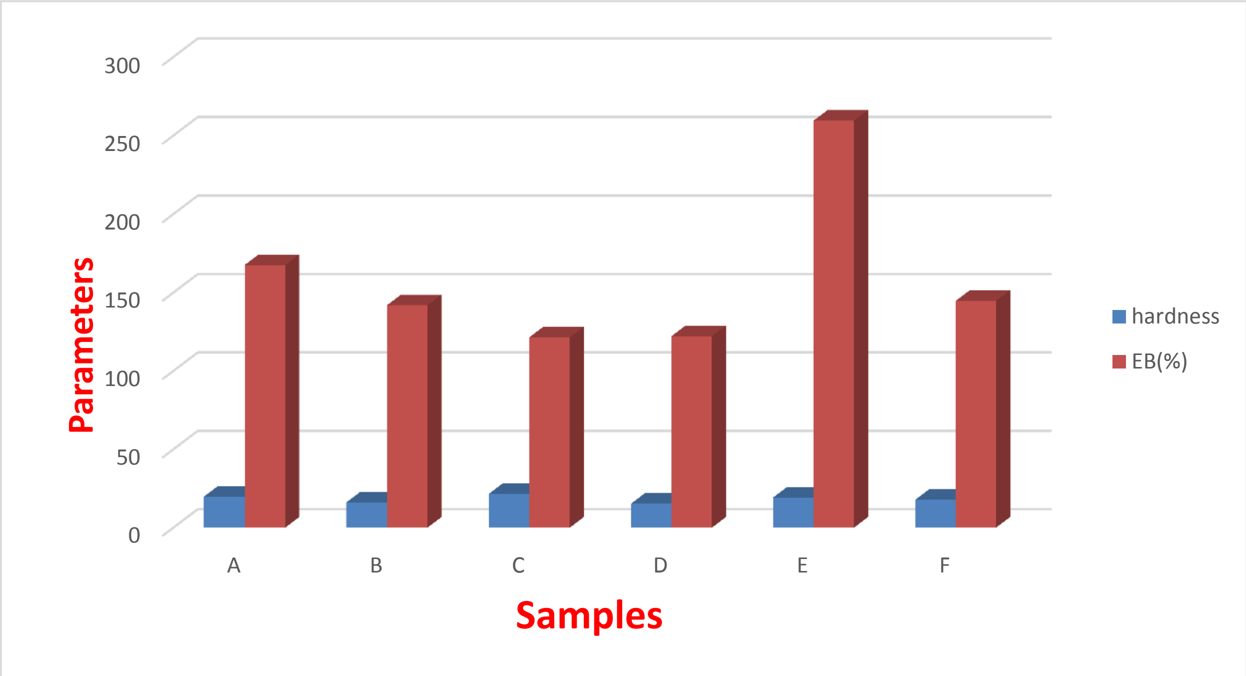

Figure 1: Mechanical properties of vulcanizates at different filler loading

The amount of RRP did not significantly influence the hardness and the abrasion resistance of the NR composites as shown in Figure 2. Small differences observed may be due to the varying composition of RRP which had been obtained from the used tires. The decrease may be due to the partially devulcanized RRP by the MBTS accelerator. The highest hardness value observed at sample C might be due to a suitable blend ratio of CB and RRP but nevertheless the blends have good hardness properties [15].

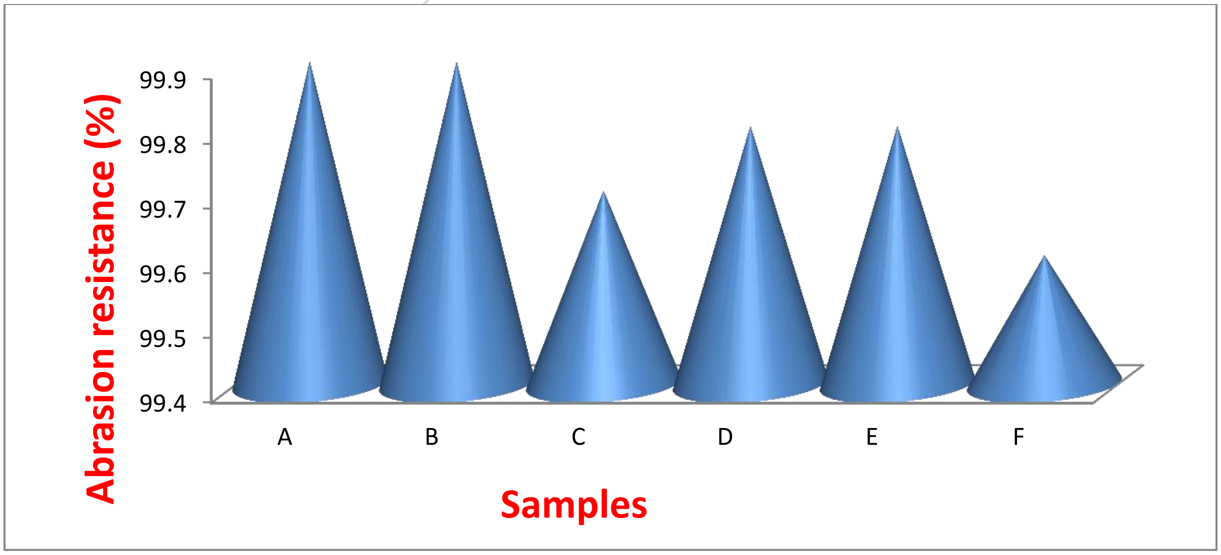

Figure 2: The abrasion resistance of vulcanizates.

The values of the ultimate tensile strength increased from C to E (Figure 3). This may be due to RRP containing remnant of sulfur and accelerator which produced reinforcing effect on the blends. Hassan *et al*. [18] reported that the increase may be due to the residual curing property of elemental sulfur and organic accelerator contained in the RRP.

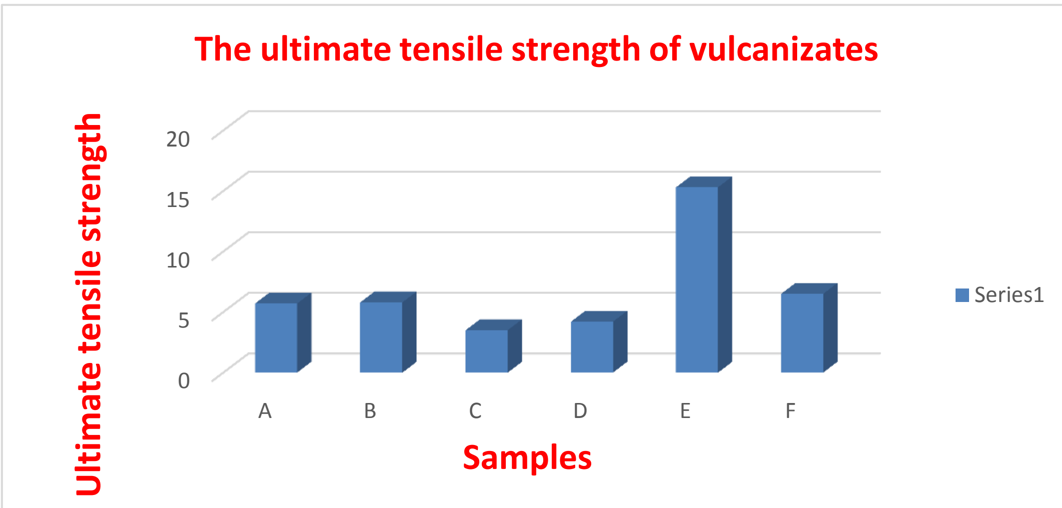

Figure 3: The Ultimate tensile strength of vulcanizates.

Formela and Haponiuk [17] suggested that it might be due to the migration of CB from the RRP into the rubber matrix which may be induced by shear forces during compounding. He also reported that the partial devulcanization of RRP increases the surface roughness and specific surface of the devulcanized RRP particles and provide better adhesion between RRP particles and the NR matrix. This was more visible at sample E because of higher NR content and consequently higher degree of devulcanization in the RRP.

3.1 Fatigue resistance

Table 3 presents the results of the fatigue test carried out on the vulcanizates.

Table 3: Result of fatigue test

Samples	Observation/ Remarks
A	No visible crack but minimal fatigue
B	No visible crack and no sign of fatigue
C	No visible crack and no sign of fatigue
D	No visible crack and no sign of fatigue
E	No visible crack and no sign of fatigue
F	No visible crack and no sign of fatigue

The results revealed that the vulcanizates containing recycled rubber powder regardless of the ratio showed complete resistance to fatigue as there were no signs of crack and fatigue in these composites. As reported earlier this might be due to the residual content of sulfur and organic accelerator in the recycled rubber powder producing reinforcing effect on the vulcanizates containing them [16]. The blend that was unloaded with recycled rubber powder showed minimal sign of fatigue but no visible crack, this result could be due to the absence of rubber powder in the composite and the crack resistance may be due to the presence of carbon black and the unique property of NR to crystallize at high strain.

Recommendations and Conclusion

The mechanical properties of most vulcanizates decreased with increased RRP loading and the abrasion and fatigue resistance of most vulcanizates showed improved values, an observation which might be due to the reinforcing effect of both fillers and the unique abrasion properties of natural rubber. However, the ultimate tensile strength increased with higher RRP content; an observation which might be due to residual content of sulphur and organic accelerator in recycled rubber powder. Waste rubber tire when properly recycled and modified can be of importance in the industries and other applications requiring the use of sulphur and organic accelerators. It is therefore recommended that waste rubber tyre be properly utilized to reduce environmental pollution and reduce the cost of sulphur and organic accelerators resulting in the overall cost benefit of the vulcanizates.

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