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Physico-Mechanical Properties and Water Absorption Behavior of Natural Rubber Vulcanizates Filled With Sawdust

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Abstract

The physical properties and water absorption behavior of vulcanized natural rubber filled with sawdust (of 60µm particle size) were investigated and compared with carbon black filled vulcanizates. The sawdust was characterized in terms of moisture content (9.80%), iodine absorption number (72.40), pH (6.40) and compounding was done at varying filler loadings (10 pphr, 20 pphr, 30 pphr and 40 pphr) using two roll mill. The results showed that incorporation of sawdust into the vulcanized natural rubber generally increased the tensile strength, tensile modulus, and hardness and abrasion resistance of the composites produced, whereas the elongation at break and compression set of composites decreased as the filler loadings increased. The results obtained from vulcanized natural rubber were compared with vulcanized rubber filled with carbon black filler. It was observed that vulcanized natural rubber filled with carbon black filler showed better mechanical properties than vulcanized natural-sawdust composite in all the filler loadings. In addition, the effects of these fillers on the end-use properties such as water absorption were examined. The results showed that as the filler loadings increase, the water uptake in sawdust filled vulcanizates increased to 5.00%, 8.40% and 9.60% for 20 pphr, 30 pphr and 40 pphr respectively showing high water absorption, while the control and carbon black filled vulcanizates had low water uptake of 0.70% for control, 2.10%, 1.30%, 0.66%, and 0.65% for 10 pphr, 20 pphr, 30 pphr, and 40 pphr respectively for carbon black filled vulcanizates.

Keywords:* *Absorption; Carbon black; Filler; Sawdust; Vulcanizates.

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Introduction

Natural rubber (NR) is an interesting material with commercial success due to its excellent physical properties, especially high mechanical strength, low heat build-up, excellent flexibility, and resistance to i Mpact and tear, and above all its

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renewability. However, raw dry rubber is seldom used in its original state for any engineering and domestic application. Consequently, rubber manufacture involves the addition to rubber many ancillary materials called additives to allow the rubber compounds to be satisfactorily processed and vulcanized in order to improve the application properties of the rubber compound [1]. Additives used in rubber manufacture include vulcanizing agents, accelerators, activators and/or retarders, fillers, anti-degradants, among others. Fillers represent one of the most important additives used in rubber compounding. Fillers are added to rubber formulation in order to optimize properties needed for service application. Reinforcement of rubber polymers with particulate fillers is a subject that had captured the interest of a large number of researchers. Property advantages obtainable from filler reinforced rubber vulcanizates include design flexibility, improved physico-mechanical properties such as tensile properties, hardness, and processing economy [2]. Due to strong environmental regulations worldwide and increased interest in the proper utilization of renewable natural resources, efforts have been made to find alternative reinforcements that are environmentally friendly while providing the same performance as their synthetic counterparts by Egwaikhide et al. With their low cost, easy availability, ease of chemical and mechanical modification, and high specific mechanical properties, natural fibres represent a good, renewable and biodegradable alternative to the most common synthetic reinforcement by Lovely et al. They achieve performance enhancement by forming strong chemical bonds with the rubber, that is, strong filler-elastomer interactions [3]. Sawdust is loose particles or wood chippings obtained as by-products from sawing of timber into standard useable size. The main chemical components of sawdust are carbon (60.8%), hydrogen (5.19%), oxygen (33.83%) and nitrogen (0.90%). In developing countries like Nigeria, proper utilization of sawmills waste has not been given due attention. The sawdust thereby constitutes an environmental nuisance as they form refuse heaps in the areas where they are disposed [4]. The use of sawdust as a partial replacement to carbon black in rubber will provide an economic use of the by-product. This research work is aimed at developing filler for rubber compounds which can be an alternative to the commonly used commercial carbon black filler with a consequent reduction in cost and may offer some new scientific knowledge and economic benefits [5].

Experimental Method

Materials and Equipment

The natural rubber used for the research work was obtained from Rubber Research Institute of Nigeria (RRIN), Iyanomoh Benin-City [6]. Sawdust was obtained from Njoku and Sons Woodmill Co Mpany, Aba-Owerri Road, Owerri North, Imo State. The sieving of the fillers was done at Technology Incubation Center National Board for Technology Incubation, Federal Ministry of Science and Technology at TIC Complex Industry Road Aba, Abia State with mesh size of 60 μm. The chemicals such as toluene; benzene and n-hexane were bought from Chemscience Laboratory at Mbaise Road by Douglas Owerri Municipal, Owerri Imo State. The rubber compounding chemicals such as processing oil, tetramethylthiuram disulphate (TMTD), mercapto benzyl thiazoledisulphate (MBTS), zinc oxide, sulphur and stearic acid were of commercial grades [7].

Equipment

The equipment used was;

i. Two roll mill, Manufactured by British Co Mpany Limited, England.
ii. Hydraulic Press, Elektron Technology Series, UK.
iii. Monsanto Tensile Tester Model (1/m) Manufactured by British Co Mpany Limited, England.

iv. Wallace Hardness Tester model C8007/25 for Hardness Test, Elektron Technology Series, UK.

v. Taber Oscillating Abrasion Tester, Model: 6160-F735, Manufactured by Taber Co. Ltd, Canada was used for the Abrasion Properties.

vi. CTM-2P-200-2000KN (200Tons), Manufactured by Interlaken Technologies Co. Ltd Thailand was used for the Compression Set.

Materials

Characteristics of filler

The sawdust was sun dried for 3days, foreign particles were removed then it was sieved with a mesh of size 60µm mesh, which was the particle of the sawdust used for the experiment. The fine particles that passed through were collected, characterized in terms of iodine value, moisture content, and pH.

Preparation of composites

The rubber was masticated and mixed with an additives using the two roll mill and adopting the standard method specified in the ASTM-D 3184-80 for all the composites. The filler loadings were varied from 0 to 40 pphr. A separate set of composites was prepared using carbon black as filler and also varied within the same range. This served as basis for co Mparison in this work. The TABLE 1 shows the formulations for the natural rubber composites. The rubber mixes were prepared on a laboratory size two roll mill. It was maintained at 70°C to avoid cross-linking during mixing after which the rubber composite was stretched out. Mixing follows [ASTMD 3184–80, 1983].

TABLE 1. **Formulations for Reinforced natural rubber composites.**

Ingredients	Part per Hundred of Rubber (pphr)
NR	100
Zinc oxide	5
MBT	1
TMTD	1
Fillers (carbon black and sawdust)	(0 - 40)
Sulphur	2.5
Processing oil	2
Stearic acid	2.5
A batch factor of two (2) was used.	

Curing of composite samples

Using Sulphur was the vulcanizing agent; the curing was done at 140°C for 5 min.

Water absorption test

The water adsorption test was carried out by cutting out samples of 20 mm × 10 mm with thickness of 5 mm. Each sample was air dried, weighed (M_1) then placed in beakers of water of 400 ml and then covered for 2 days at a room temperature (32°C). After which they were taken out, wiped with a filter paper, re-weighed (M_2). The water absorption was calculated using the equation.

$$\text{Water Absorption} = \frac{M_2 - M_1}{M_1} \times 100$$

Results and Discussion

The results are discussed as below:

TABLE 2. **Filler characteristics.**

Sample Results	Sawdust (SD)	Carbon Black (CB)
pH value of 10% solution	6.4	6.6
Moisture content (%)	9.8	2.32
Iodine adsorption value	72.4	80.53
Particle size	60.00 (μm)	30.0 - 32.0 (nm)

TABLE 3. **Mechanical properties of vulcanizates.**

Property	Filler Loadings (pphr)				
	CT	10	20	30	40
Tensile Strength (Mpa)	10.25	(20.45), [18.58]	(25.25), [20.15]	(29.68), [23.55]	(33.05), [27.11]
Elongation (%)	803	(650), [735]	(502), [695]	(475), [601]	(379), [515]
Tensile Modulus (Mpa)	1.68	(2.50), [1.95]	(2.93), [2.05]	(3.54), [2.48]	(3.93), [3.04]
Hardness (IRHD)	35.03	(44.56), [41.69]	(47.23), [43.50]	(50.15), [44.75]	(56.24), [48.06]
Abrasion Resistance (%)	18.5	(24.18), [25.46]	(28.25), [29.82]	(33.61), [34.29]	(34.15), [36.35]
Compression Set (%)	25.13	(16.06), [19.58]	(12.42), [17.26]	(10.01), [14.19]	(8.24), [11.50]
Keys: Carbon black in ( ) Sawdust in [ ] pphr = Part per hundred of rubber					

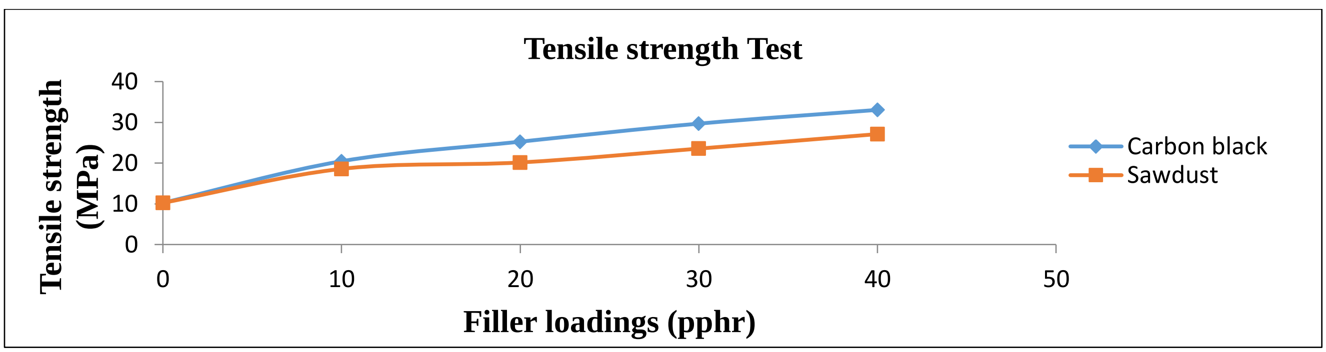

FIG. 1. **Effect of filler loadings on tensile strength of filled NR vucanizates.**

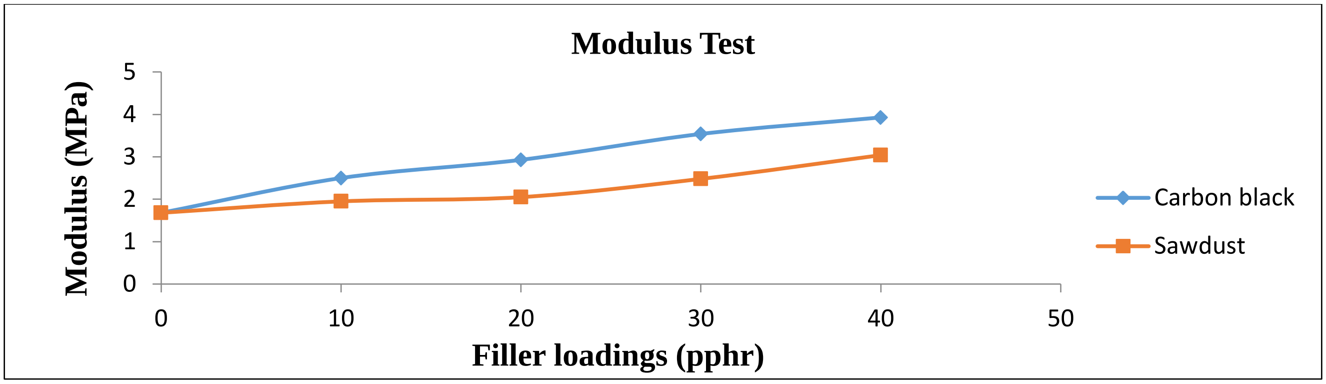

FIG. 2. **Effect of filler loadings on tensile modulus of filled NR vucanizates.**

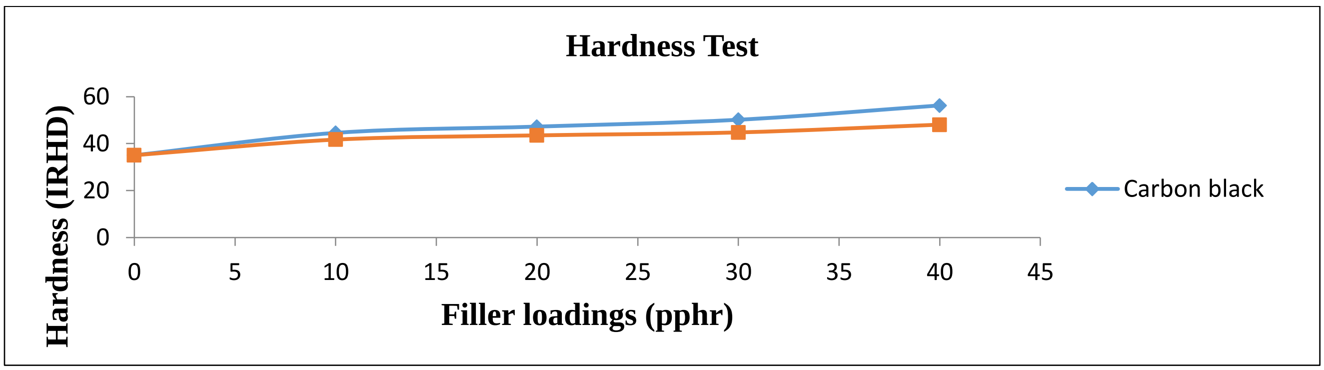

FIG. 3. **Effect of filler loadings on hardness of filled NR vucanizates.**

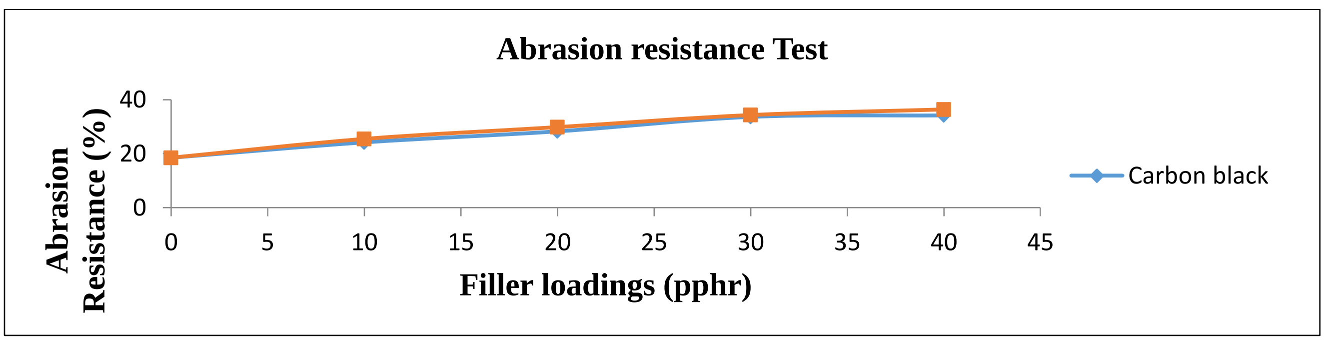

FIG. 4. **Effect of filler loadings on abrasion resistance of filled NR vucanizates.**

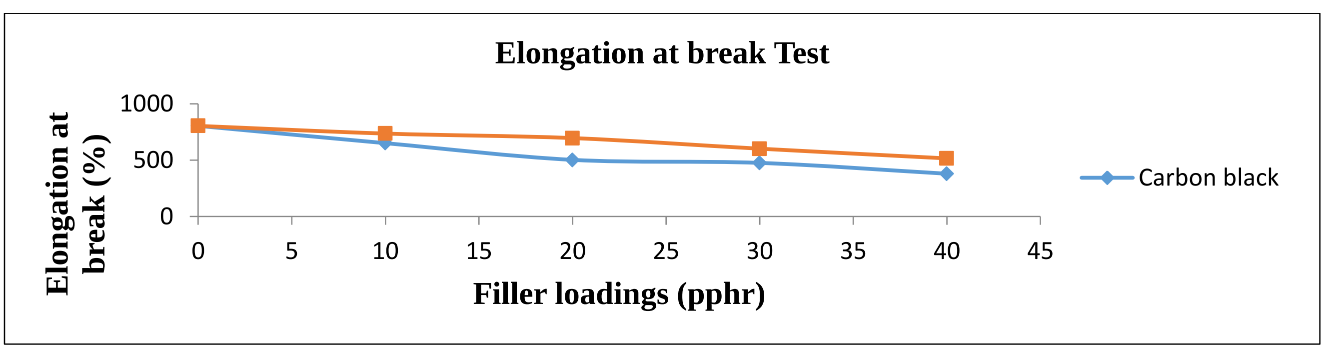

FIG. 5. **Effect of filler loadings on elongation at break of filled NR vucanizates.**

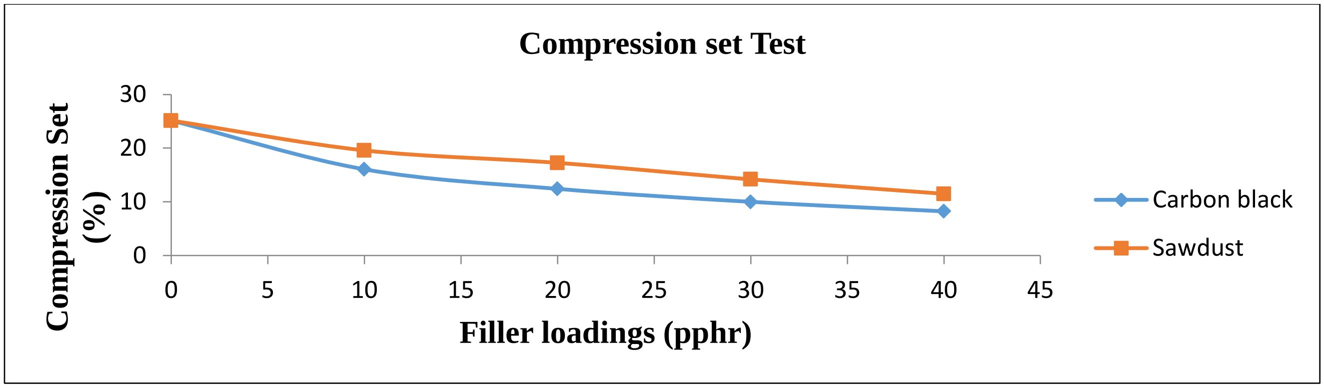

FIG. 6. **Effect of filler loadings on compression set of filled NR vucanizates.**

TABLE 4. **Water absorption.**

Matrix	Filler loading (pphr)	Water absorption (%)
CT	0	0.7
Carbon black	10	2.1
	20	1.3
	30	0.66
	40	0.65
Sawdust	10	2.9
	20	5
	30	8.4
	40	9.6

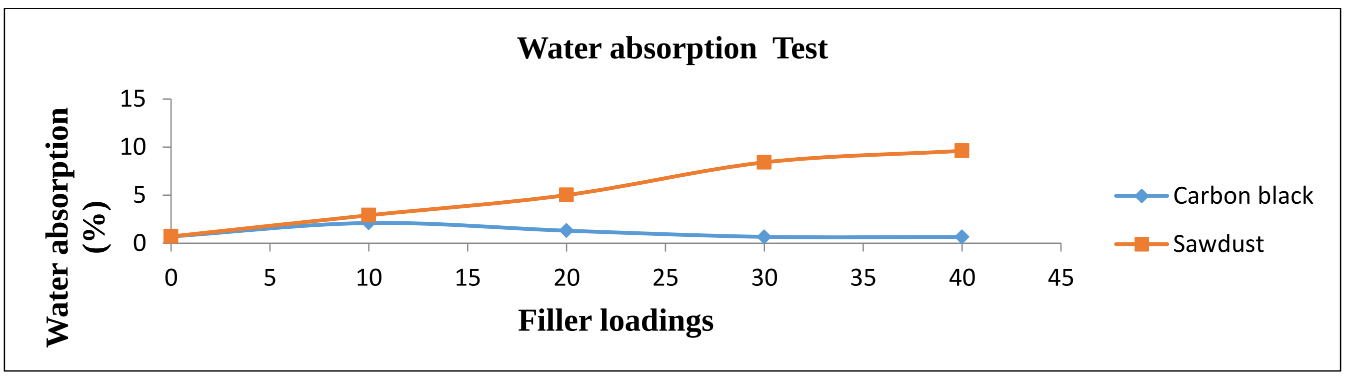

FIG. 7. **Effect of filler loadings on water absorption of filled NR vulcanizates.**

Discussion

Result of filler characteristics

The iodine adsorption value is a measure of the surface area of the filler, the higher it is, the finer or smaller the particle size of the filler and the more the reinforcement potential (Blow et al, 1982 and Hepbum, 1984). The TABLE 2 showed that iodine adsorption value of carbon black was higher than sawdust, indicating that the carbon black has a better reinforcing property than sawdust. The moisture content of fillers obtained were 9.80% and 2.32% for sawdust and carbon black respectively. The lower the moisture content, the lower the degree of defect arising from shrinkage during curing at elevated temperature (Egwaikhide et al, 2007). The pH values of the fillers were slightly acidic. The pH at acidity level tends to slow cure rate and hence reduce the cross-link density (Mohanty el at, 2001). The particle size of carbon black was finer than the sawdust as shown on the TABLE 2. It is believed that the finer the particle size, hence large surface area, the greater the interaction between the filler and rubber matrix [8].

Result of mechanical properties

In the filled system as shown in (FIG. 1-4) and TABLE 3 showed that tensile strength, modulus, hardness and abrasion resistance increased as the filler loading increased [9]. When the filler loading was 10 pphr, the tensile strength, modulus, hardness and abrasion resistance for carbon black filled vulcanizates were 20 Mpa, 2.50 Mpa, 44.56IRHD and 24.18% respectively, for sawdust filled vulcanizates were 18.58 Mpa, 1.95 Mpa, 41.69IRHD and 25.46% respectively but when the filler loading increased to 40 pphr, the tensile strength, modulus, hardness and abrasion resistance for carbon black filled vulcanizates were 33.05 Mpa, 3.93 Mpa, 56.24IRHD and 34.15% respectively and 27.11 Mpa, 3.04 Mpa, 48.06IRHD and 36.35% respectively for sawdust filled vulcanizates [10]. The results agreed with results obtained for rice husk filled coconut fiber and palm kernel husk. The carbon black used as reference filler in this study steadily showed significant increases in the tensile strength, modulus and hardness, this may be attributed to high carbon content in carbon black. The effectiveness of filler may be measured by its carbon content. Fillers with higher carbon content, provide greater reinforcement than those with lower carbon content because carbon itself is very good reinforcing filler (Okieimen et al, 2003). It is also expected that the modulus and hardness will increase because as more filler particles get into the rubber, the elasticity of the rubber chain is reduced, resulting in more rigid vulcanizates [11]. The results of tensile strength, modulus and hardness of carbon black filled vulcanizates were higher than the sawdust filled vulcanizates in all filler loadings for this reason; sawdust filler show a greater tendency towards filler agglomeration, making dispersion and distribution more difficult. The abrasion resistance of sawdust filled vulcanizates was higher than carbon black filled vulcanizates, because of distribution of carbon black into the polymer matrix [12].

TABLE 3 and FIG. 5 showed that the elongation at break decrease with increasing filler loadings. The carbon black filled vulcanizates showed a higher elongation at break than sawdust filled vulcanizates at all filler loadings, the unfilled having the highest elongation at break [13]. The observation was also found to be in agreement with the observations made by some other workers. For instance, with partially ashed rice husk and completely ashed rice husk (WRHA) (Ishak and Bakar, 1995), rubber seed shell carbon (RSSC). In general, the incorporation of reinforcing or non-reinforcing (inert) fillers into natural rubber produces decreases in elongation at break of rubber vulcanizates. The decreasing trend in elongation at break with increasing filler loading is attributed to increase in stiffness and brittleness, which decreased the resistance to stretch in application of strain [14]. The elongation at break for carbon black filled vulcanizates and sawdust filled vulcanizates for 10 pphr is 650% and 735% respectively while for 40 pphr were 379% and 515% respectively.

TABLE 3 and (FIG. 6) showed the compression decreased with increase in all filler loadings [15]. The lower values of the results indicate the best material for the compression state and high compression indicates less reinforcement [16]. The decrease is expected because as filler loading concentration increased in the polymer matrix, the void space in the matrix is reduced, hence there is decrease in percentage compression (Abode, 2010). The unfilled vulcanizate had the highest compression set, the compression set of sawdust filled vulcanizates of all the filler loadings were higher than that of carbon black vulcanizates [17]. This indicating that the carbon black filled vulcanizates had the best compression state at highest filler loading (40 pphr), compression set for carbon black filled vulcanizate was 8.24% co Mpared to sawdust filled which had 11.50% and at low filler loading (10 pphr), carbon black filled was 16.06% co Mpared to sawdust filled with 19.58% [18].

Result of water absorption

From TABLE 4 and (FIG. 7) of water absorption, it is expected that water absorption level would decrease with filler loading, but the water uptake varied among the two fillers. For carbon black filler loading 10phr had 2.10% and 20% had 1.30%, these might be due to voids between carbon black and the rubber matrices that allowed much water uptake. The water uptake reduced as the filler loading increased because as the filler increases in the rubber matrix, the voids between carbon black and rubber matrix tends to reduce [19]. The filler loading of 40 pphr had the lowest water absorption for carbon black vulcanizate. The sawdust filled vulcanizates showed an increase in water absorption as filler loading increased. Sawdust being a cellulosic in nature, has some hydroxyl groups, thereby make it hydrophilic (water loving) in nature (Tenebe et al, 2013), which are able to form hydrogen bond between water and sawdust. As the filler loading increased, the number of hydrogen bonds between organic components and water molecules also increased. This free hydroxyl (OH) groups in the sawdust come in contact with water through hydrogen bonding, which results in water uptake and weight gain in the composite. Another reason that might have increased water absorption in sawdust filled vulcanizates [20], might be poor filler dispersion and filler-polymer matrix interaction between the sawdust and natural rubber

Conclusion

This study showed that incorporation of carbon black and sawdust into natural rubber vulcanizates increases the tensile strength, tensile modulus, hardness and abrasion resistance with increase in filler loading, whereas the elongation at break, compression set and flex fatigue decreases as filler loading increases [21]. The fillers under investigation (carbon black and sawdust) have shown their effects on the vulcanizates produced at different filler loadings. Hence, the mechanical properties of the composites produced are found to be a function of matrix filler adhesion, dispersion of fillers within the matrix and particle size. It could be foreseen that these properties could make these composites produced desirable for some applications, such as shoe sole, foot mat, footwear, flooring, especially at filler loading (30 pphr and 40 pphr). The sawdust filled vulcanizates investigated showed significant increase in water absorption as the filler loading increased than carbon black filled vulcanizates. The sawdust filled vulcanizates showed higher water absorption, filler loadings of 30 pphr and 40pphr having the higher values of 8.40% and 9.60% respectively due to higher presence of hydroxyl (OH) group in sawdust. The best filler loadings required for best applications is 30 pphr and 40 pphr for carbon black filled vulcanizates because they had lowest water absorption of 0.66% and 0.65% respectively [22].

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