MECHANICAL PROPERTIES AND WATER ABSORPTION BEHAVIOR OF NATURAL RUBBER FILLED WITH MELON HUSK AND SAWDUST

Amoke, A.
Department of Polymer Technology,
Auchi Polytechnic Auchi, Edo State, Nigeria
E-mail address: amokeaustine@yahoo.com

Onyegbule, U. D.
Department of Chemical Engineering Technology,
Auchi Polytechnic, Auchi Edo State, Nigeria

Anetokhe, M.A.
Department of Chemical Engineering Technology,
Auchi Polytechnic, Auchi Edo State, Nigeria

Atulute O. O.
Department of Chemical Engineering Technology,
Auchi Polytechnic, Auchi Edo State, Nigeria

ABSTRACT

Fillers are materials which when added to rubber mix enhance the properties. These properties are physical in nature which include hardness, tensile strength, flex fatigue, stiffness and to some extent, the chemical properties. Fillers improve the processing characteristics, reduce cost and also act as auxiliary components necessary for vulcanisate. In rubber industry, fillers that are commonly in use are carbon black, China clay and calcium carbonate. The process of producing carbon black requires tremendous energy utilization, heavy infrastructure setup and constitutes a heavy source of environmental pollution and it is also carcinogenic. Nowadays, there has been a growing interest in the use of individual and agricultural waste as fillers because of their renewability, low cost, readily available and environmental friendly. The mechanical properties and water absorption behavior of natural rubber filled with melon husk and sawdust as fillers were studied. The melon husk and sawdust were characterized in terms of moisture content (melon husk = 5.80% and sawdust = 9.80%), iodine absorption number (melon husk = 59.00 and sawdust = 70.40), pH (melon husk = 6.30 and sawdust = 6.40), particle size of the fillers were 75µm and compounding was done at varying filler loadings (10 to 40pphr) using two roll mill machine. The mechanical properties result showed that incorporation of melon husk and sawdust into the vulcanized natural rubber generally increased as filler loadings increased. The highest tensile strength (sawdust = 10.25-27.11 MPa and melon husk = 10.25-25.03 MPa), tensile modulus (sawdust = 1.68-3.04 MPa and melon husk = 1.68-2.35 MPa), hardness (sawdust = 35.03-48.06 IRHD and melon husk = 35.03-45.92 IRHD) and abrasion resistance (sawdust = 18.50-36.35% and melon husk = 18.50-30.13%) were obtained as filler loadings increased. The elongation at break (sawdust = 803-515% and melon husk = 803-560%), compression set (sawdust = 25.13-11.50% and melon husk = 25.13-12.75%) and flex fatigue (sawdust = 15.45-7.29 Kc x 10^3 and melon husk = 15.45-7.85 Kc x 10^3) of composites decreased as the filler loadings increased. The sawdust filled vulcanizates showed better mechanical properties than melon husk filled vulcanizates. 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 increased, the water uptake in melon husk filled vulcanizates and sawdust filled vulcanizates increased due to hydrophilic nature of the fillers. The sawdust filled vulcanizates had more water uptake than the melon husk filled vulcanizates.

Keywords: Absorption, Filler, Melon husk, Natural rubber, Sawdust

INTRODUCTION

Filler is one of the major additives used in natural rubber compound and has marked effect and influence on rubber materials. Filler play a dominant role in modifying the physical properties of base polymer. (Thomas et al, 2013). Fillers are materials which when added to rubber mix enhance the properties. These properties are physical in nature which include hardness, tensile strength, flex fatigue, stiffness and to some extent, the chemical properties. Fillers improve the processing characteristics, reduce cost and also act as auxiliary components necessary for vulcanisate. Fillers can either be reinforcing, semi-reinforcing or non-reinforcing. Reinforcing fillers enhance the physical properties of the cured article (Okieimen and Imanah, 2003). There are also non-reinforcing fillers, they reduce cost. Non-reinforcing fillers have little or no effect on the physical properties of the rubber. Examples of these include talc, barites, mica powder, whiting and china clay. Semi-reinforcing fillers are partially reinforcing. These include soft clay, calcium carbonate and antimony (Marut et al, 2015; Okwele et al, 2018). In rubber industry, fillers that are commonly in use are carbon black, China clay and calcium carbonate. The process of producing carbon black requires tremendous energy utilization, heavy infrastructure setup and constitutes a heavy source of environmental pollution and it is also carcinogenic (Okwele et al, 2018). Nowadays, there has been a growing interest in the use of individual and Agricultural waste such as rice husk (Attharangsan et al., 2012). Agricultural by-products as fillers has been investigated, this included banana peel, rice husk, spent mango, bean seed skin and groundnut shell (Tenebe et al., 2019), melon and sawdust (Amoke et al, 2017). Agricultural by-products are low cost materials and readily available in large quantity for use everywhere, of which well over 300 million tones are produced annually (Okwele et al, 2018).

OBJECTIVES

i. To compare the mechanical properties of natural rubber filled with sawdust and melon husk..
ii. To study the water absorption behaviour of the natural rubber filled with sawdust and melon husk.

MATERIALS AND METHODS

Materials

Nigeria standard rubber of grade NSR-10 used for the research work was obtained from Rubber Research Institute of Nigeria (RRIN), Iyanomoh Benin-City. Sawdust was obtained from Njoku and Sons Woodmill Company, Aba-Owerri Road, Owerri North, Imo State. Melon husk was obtained from Oban local Market, Nsukka, Enugu 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.

METHODS

Preparation of Fillers

The melon husk was soaked in water for one hour for heavy particles, such as; stones, metals, mixed with it may settle at the base and the melon husk was collected from the top surface of the water since they were lighter. It was then dried under sun. Some foreign particles that were light were also removed. The melon husk was then ground into fine powder using an automated grinding machine. The sawdust was sun dried and foreign particles were also removed. It was not ground because it was collected as fine powder from wood shelving. Both melon husk and sawdust were then sieved with a mesh of size of 75µm mesh.

Fillers Characterization

In the course of this work, it was really necessary to characterize the fillers which were the most important of the study, to enhance identification. The parameters of the fillers which were determine included moisture content, pH value, particle size and iodine absorption value.

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. The Table 1 shows the formulations for the natural rubber composites. The rubber mixes were prepared on a laboratory size two roll mill. Its temperature was maintained at 70^0C to avoid cross-linking during mixing after which the rubber composite was stretched out. Mixing follows [ASTMD 3184–80, 1983].

Table 1: Formulation for the Filled Natural Rubber composites

Ingredients	Part per Hundred of Rubber (pphr)
NR	100
Zinc oxide	5.0
Stearic acid	2.5
(MBTS)	1.0
(TMTD)	1.0
Fillers (sawdust and melon husk)	(0 – 40)
Sulphur	2.5
Processing oil	2.0

Mechanical Properties of the Composite

The mechanical properties of the composites were determined using standard test procedures. The tensile properties determination was carried out in accordance with ASTMD 412-87 method. Hardness Test was done in accordance with ASTMD 785. ASTMD 385 method was used for compression set determination. The flex fatigue determination was carried out using ASTMD 430 using the flex tensometer machine. Wallace Akron abrasion tester was used for abrasion resistance determination.

Water Absorption Test

The water adsorption test was carried out by cutting out samples of 20mm x 10mm x 5mm. Each sample was air dried, weighed (M_1) then placed in different beakers of water 400ml and then covered for 2 days at a room temperature (32^0C). After which they were taken out, wiped with a filter paper, re-weighed (M_2). The water absorption was calculated using the equation.

$$\text{Water absorption} = \frac{M_2 - M_1}{M_1} \times 100 \qquad (1)$$

RESULTS AND DISCUSSION

Results

Table 2: Fillers Characterization

Sample Results	Melon Husk	Sawdust
pH value of 10% solution	6.30	6.40
Moisture content (%)	5.80	9.80
Iodine value	59.00	70.40
Particle size (µm)	75.00	75.00

Mechanical Properties of the Vulcanzates

Table 3: Mechanical Properties

Property	Filler Loadings (pphr)				
	Control	10	20	30	40
Tensile Strength (MPa)	10.25	(18.58), [17.26]	(20.15), [18.78]	(23.55), [22.05]	(27.11), [25.03]
Elongation at break (%)	803	(735), [743]	(695), [702]	(601), [643]	(515), [560]
Tensile Modulus (MPa)	1.68	(1.95), [1.80]	(2.05), [1.98]	(2.48), [2.13]	(3.04), [2.35]
Hardness (IRHD)	35.03	(41.69), [38.12]	(43.50), [42.43]	(44.75), [43.18]	(48.06), [45.92]
Abrasion Resistance (%)	18.50	(25.46), [22.03]	(29.82), [25.01]	(34.29), [28.99]	(36.35), [30.13]
Compression Set (%)	25.13	(19.59), [20.47]	(17.26), [18.63]	(14.19), [14.86]	(11.50), [12.75]
Flex Fatigue (kc x 10^3)	15.45	(12.19), [13.23]	(11.73), [12.01]	(8.25), [9.88]	(7.29), [7.85]

Key: Sawdust = ( ) Melon husk = [ ] Pphr = parts per hundred of rubber

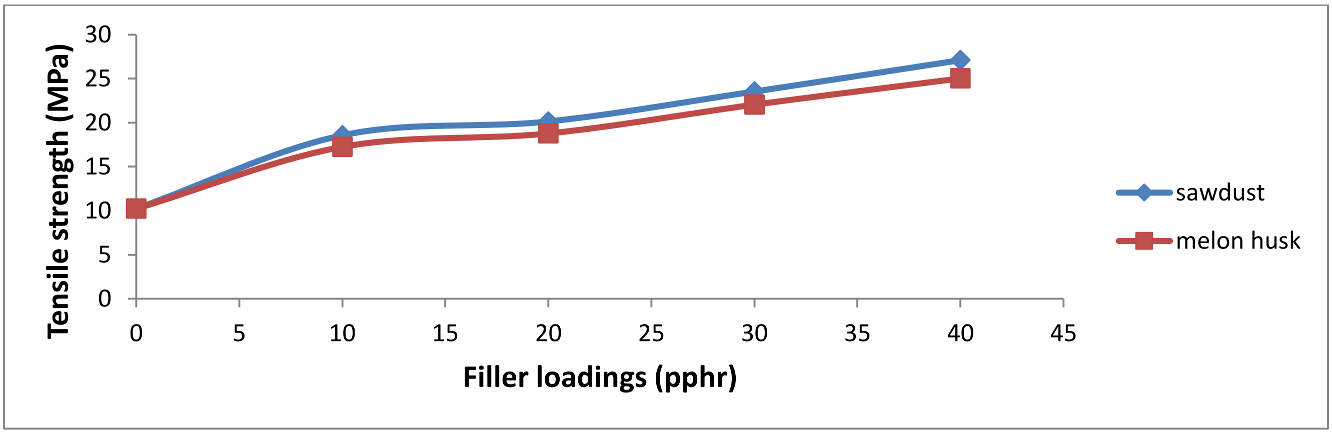

Figure 1: Effect of Filler Loadings on Tensile Strength of Filled Vucanizates Natural rubber

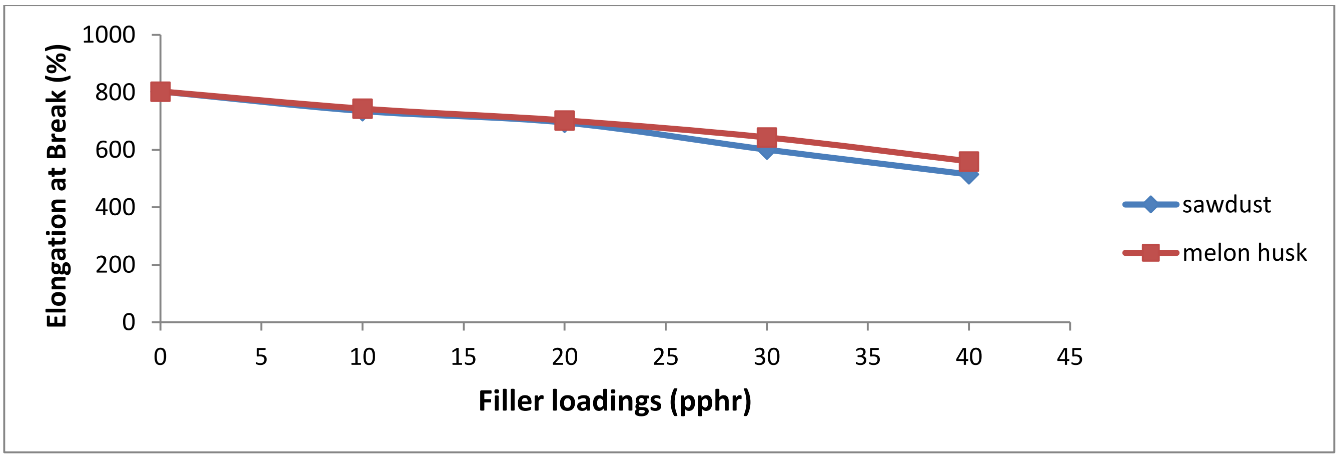

Figure 2: Effect of Filler Loadings on Elongation at Break (%) of Filled Vulcanizates Natural rubber

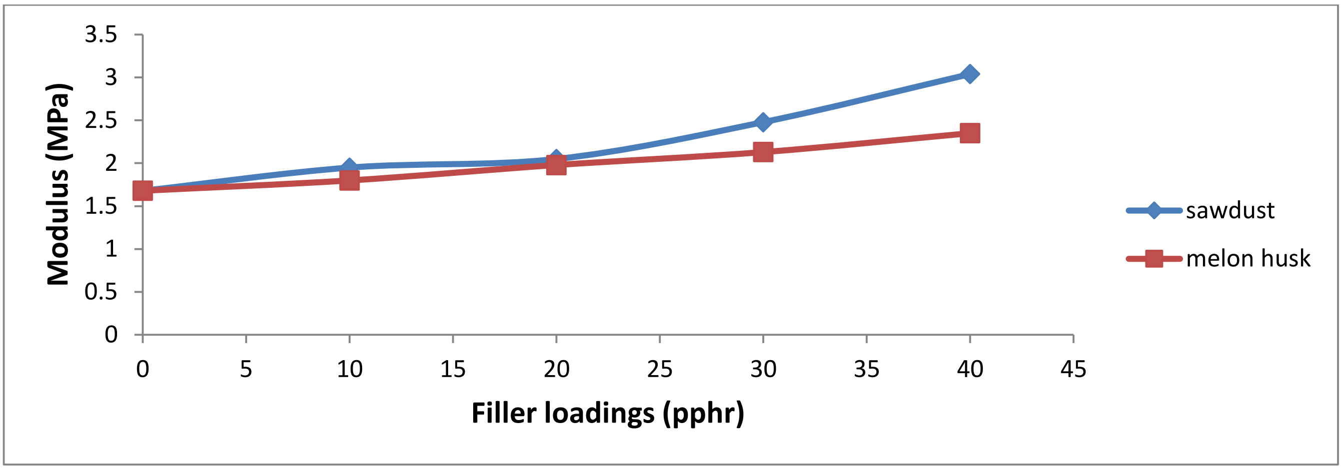

Figure 3: Effect of Filler Loadings on Modulus of Filled Vulcanizates Natural rubber

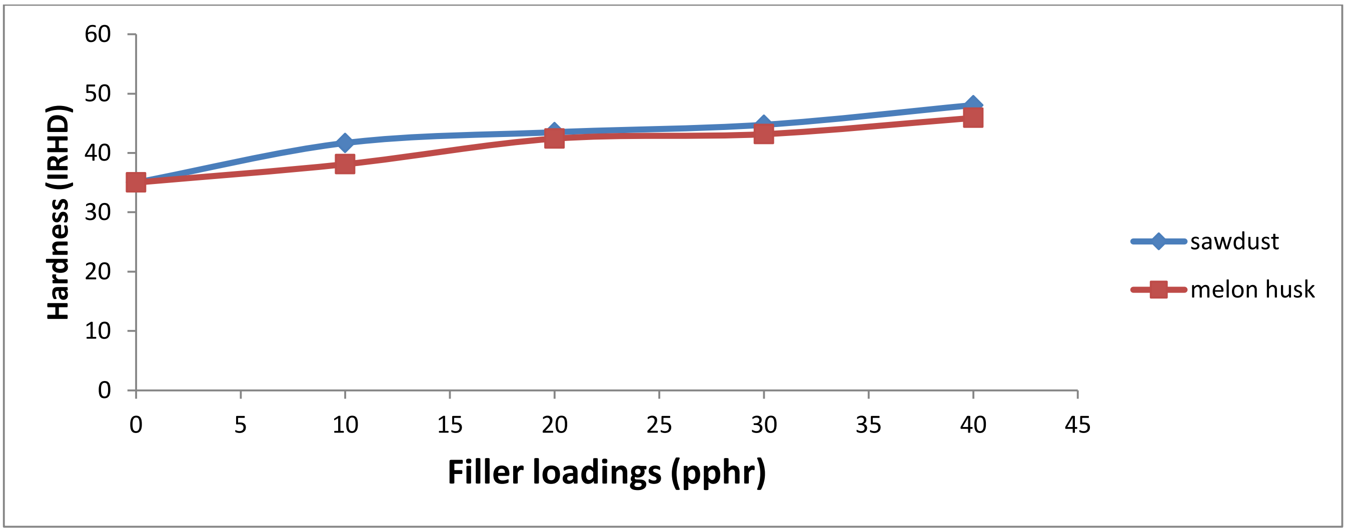

Figure 4: Effect of Filler Loadings on Hardness of Filled Vulcanizates Natural rubber

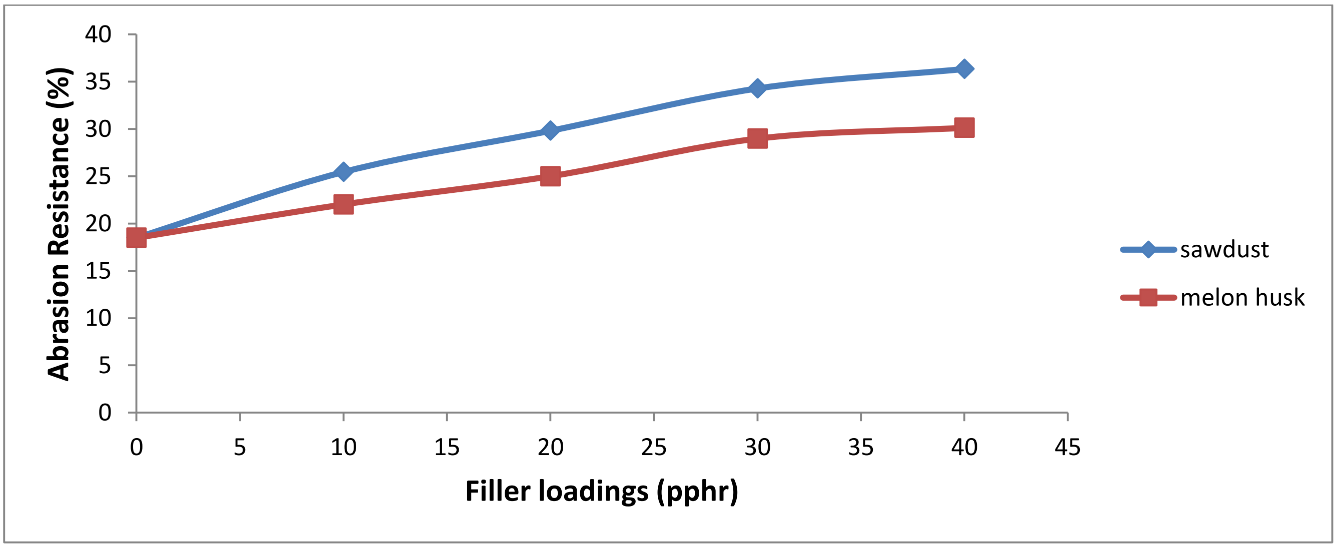

Figure 5: Effect of Filler Loadings on Abrasion Resistance of Filled Vulcanizates Natural rubber

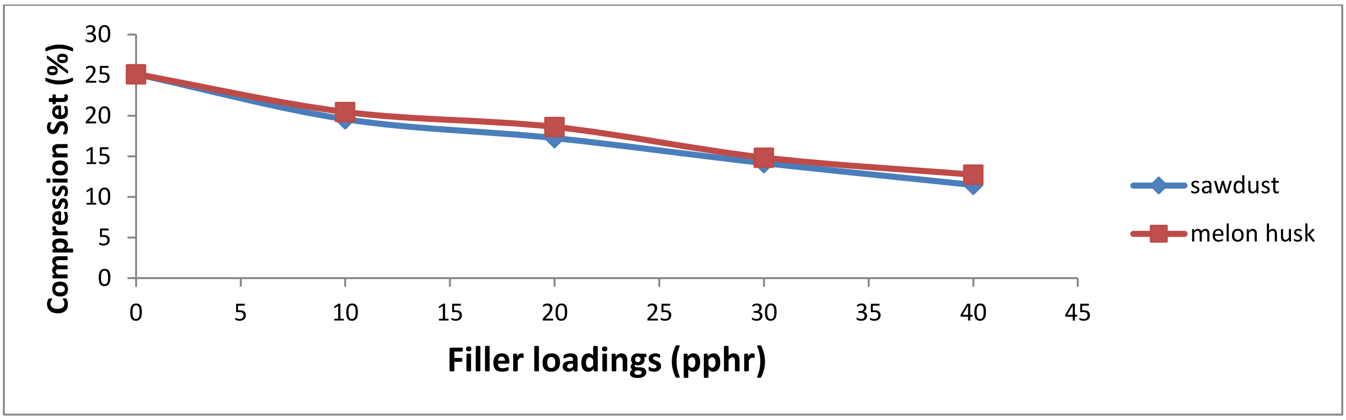

Figure 6: Effect of Filler Loadings on Compression Set of Filled Vulcanizates Natural rubber

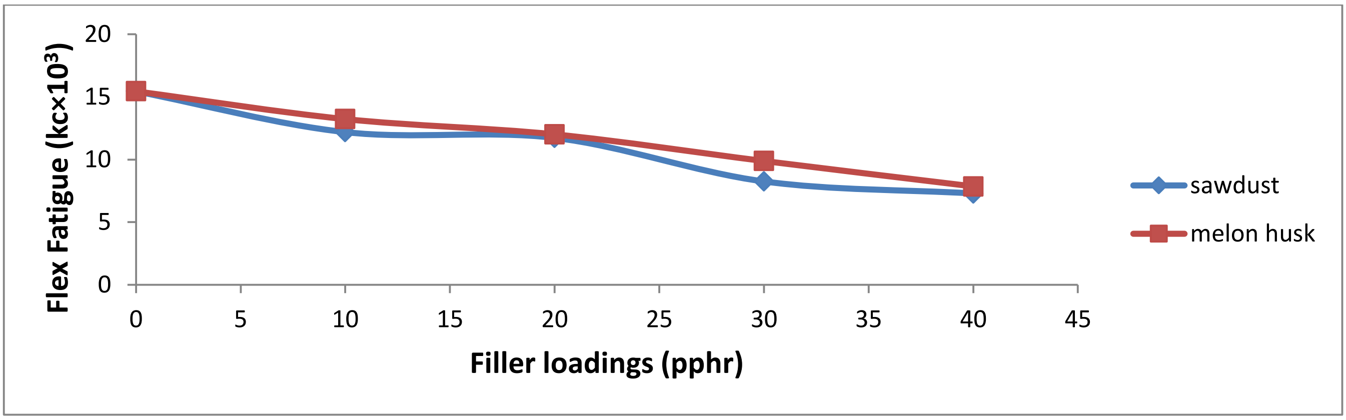

Figure 7: Effect of Filler Loading on Flex Fatigue of Filled Vulcanizate Natural rubber

Table 4: Water Absorption

Matrix	Filler loading (pphr)	Water absorption (%)
Control	0	0.70
Melon seed shell	10	2.00
	20	3.40
	30	3.90
	40	3.90
Sawdust	10	2.90
	20	5.00
	30	8.40
	40	9.60

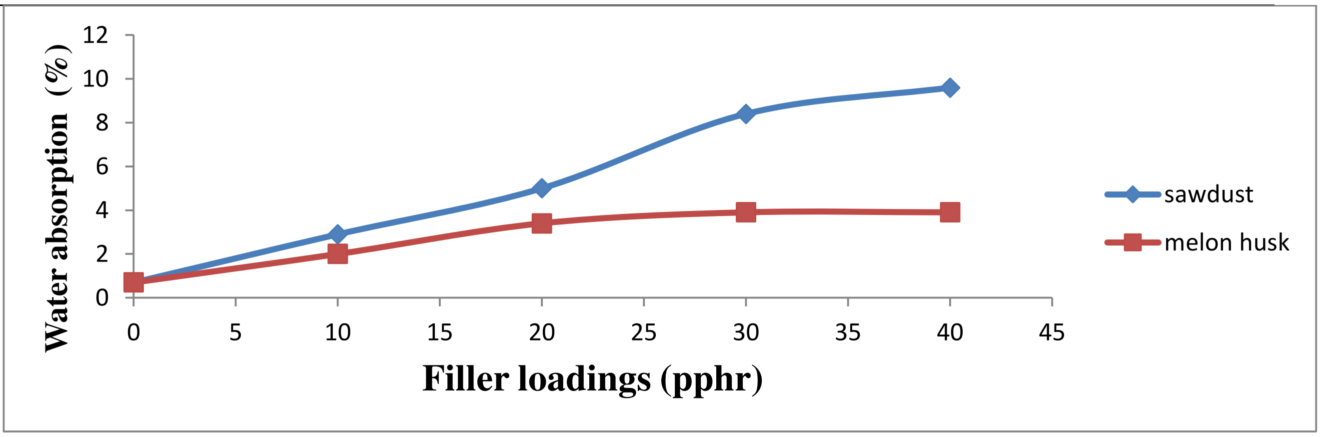

Figure 8: Effect of Filler Loadings on Water Absorption of Filled Vulcanizates Natural rubber

DISCUSSION

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 sawdust was higher than melon husk, indicating that the sawdust has a better reinforcing property than melon husk. The moisture content of fillers obtained were 9.80% and 5.80% for sawdust and melon husk. 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 as shown on Table 2. The pH at acidity level tends to slow cure rate and hence reduce the cross-link density (Mohanty et al, 2001).

Mechanical Properties

Table 3 and Figure 1 showed that tensile strength increased with increase in filler loading for all the cases considered. The increase in tensile is as a result of high surface area of the fillers, which created a better filler-rubber matrix interaction and hence a better tensile properties for the filled composites (Hussain et al, 2010 and Okiemen et al, 2002). Generally, the smaller the particle sizes of filler, the greater the tensile strength of the vulcanizates (Ismail et al, 1997). The sawdust filled vulcanizates had the better tensile strength than the melon husk filled vulcanizates.

It is also expected that the modulus and hardness will increase because as more filler particles get into the polymer matrix as seen on Table 3, Figures 3 and 4, This suggests that the incorporation of these fillers can effectively reduce the mobility of rubber chains and the stiffness of the resulting composites are increased (Anyaporn et al, 2017). The results of modulus and hardness of sawdust filled vulcanizates were higher than the melon husk filled vulcanizates in all filler loadings because there was a better filler-rubber matrix interaction between the sawdust-rubber matrix than in melon husk-rubber matrix.

Table 3 and Figure 2 showed that the elongation at break decreased with increasing filler loading. The reduction of elongation at break is due to stiffening of the polymer matrix by the filler. Further increase in filler loading causes the molecular mobility decrease due to extensive formation of physical bond between the filler particles and the polymer chain that stiffen the matrix and decreasing stretching (Sau et al, 1999). The sawdust filled vulcanizates showed a higher elongation at break than melon husk filled vulcanizates at all filler loadings. In general, the incorporation of reinforcing or non-reinforcing (inert) fillers into natural rubber produces decreases in elongation at break of rubber vulcanizates (Okieimen et al, 2003).

Thus, the abrasion resistance of a solid body is defined as its ability to withstand the progressive removal of the material from its surface as a result of the mechanical action of rubbing, scraping or erosive nature (Amoke et al, 2021). The result of abrasion resistance presented in Table 3 and Figure 5 showed that as the filler loading increased for both fillers, the abrasion resistance increased showing a better abrasion resistance. This observation may be attributed to the degree of dispersion of sawdust and melon husk particles within the matrix. The abrasion resistance of sawdust filled vulcanizates was higher than melon husk filled vulcanizates, showing that sawdust filled vulcanizates have a better resistance to surface scratch when compared to melon husk filled vulcanizates.

Table 3 and Figure 6 showed the compression decreased with increase in all filler loadings. The lower values of the results indicate the best material for the compression state and high compression indicates less reinforcement. 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 compression set of sawdust filled vulcanizates of all the filler loadings were lower than that of melon husk filled vulcanizates. This indicates that the sawdust filled vulcanizates had a better degree of filler dispersion in the rubber matrix than the melon husk filled vulcanizates.

The value of flex fatigue decreased with increasing filler loading as shown on Table 3 and Figure 7. A decrease in flex fatigue has been explained in terms of adherence of the filler to the polymer phase leading to the stiffening of the polymer chain and hence resistance to stretch when strain is applied (Ayo et al, 2010). As filler loading increases, it could be expected that more filler particles and aggregates will not be dispersed and wetted efficiently by the rubber matrix (Sukru et al, 2008).

Water Absorption

Table 4 and Figure 8 of water absorption, it is expected that water absorption level would decrease with filler loading, but the water absorption increased for both melon husk and sawdust filled vulcanizates. Sawdust and melon husk are cellulosic in nature, they have some hydroxyl groups, thereby making the fillers hydrophilic (water loving) in nature (Tenebe et al, 2013), which are able to form hydrogen bond between water and fillers (melon husk 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 and melon husk fillers come in contact with water through hydrogen bonding, which results in water absorption and weight gain in the composites. The sawdust filled vulcanizates had higher water absorption than melon husk filled vulcanizates in all the filler loadings.

CONCLUSION

The fillers under investigation (sawdust and melon husk) have shown their effects on the mechanical properties and water absorption behavior on the vulcanizates produced at different filler loadings. This study show that incorporation of sawdust and melon husk into natural rubber vulcanizates increases the tensile strength, tensile modulus, hardness and abrasion resistance with increase in filler loadings, whereas the elongation at break and compression set decreases as filler loading increases. 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, especially at filler loadings (30 pphr and 40 pphr) for sawdust filled vulcanizates which had a better mechanical properties than melon husk filled vulcanizates. The melon husk and sawdust filled vulcanizates investigated showed significant increase in water absorption as the filler loading increased due to their cellulosic nature. The sawdust filled vulcanizates showed higher water absorption, filler loadings of 30 pphr and 40 pphr had the higher values of 8.40% and 9.60% respectively than melon seed shell which had 3.90% for both filler loadings at 30 and 40 pphr.

REFERENCES

1. Abode, S.I (2010): An Investigation into the use of Carbonized Bagasse filler in rubber products. Postgraduate Diploma in Production Engineering Thesis. University of Benin, Nigeria.
2. Amoke, A., Ogbobe, O., Tenebe, G.O., Ichetaonye, S.I. and Ayo, M.D. (2017): Physico Mechanical Properties And Water Absorption Behaviour Of Natural Rubber Vulcanizates Filled With Sawdust. Research and Reviews in Polymers. 8(1):108.
3. Amoke, A; Tenebe, O.G; Ichetaonye, S.I, Edegbe, O.C and Ayo, M.D (2021): Comparison of Mechanical Properties of Natural Rubber Vulcanizates Filled with Hybrid Fillers (Carbon Black/Palm Kernel Shell and Palm Kernel Shell/Sandbox Seed Shell), International Journal of Research and Innovation in Applied Science (IJRIAS), Volume VI, Issue 1, ISSN 2454-6194

4. Anyaporn Boonmahitthisud, Peeraphong Pokphat, Phasawat Chaiwutthinan and Saowaroj Chuayjuljit (2017): Nanocomposites of NR/SBR Blend Prepared by Latex Casting Method: Effects of Nano-TiO_2 and Polystyrene-Encapsulated Nano-TiO_2 on the Cure Characteristics, Physical Properties, and Morphology Hindawi Journal of Nanomaterials Volume 2017, Article ID 7676158, 11 pages
5. Attharangsan, S., Ismail, H., Abu Bakar, M., and Ismail, J. (2012): "Carbon black (CB)/ rice husk powder (RHP) hybrid filler- filled natural rubber composites effect of CB/RHP ratio on property of the composites," Polymer-Plastics Technology and Engineering, vol. 51, issue 7, pp. 655662.
6. Ayo, M.D., Madufor, I.C., Ekebafe, L.O., Chukwu, M.N. (2010): Effects of Carbonization Temperature on the Mechanical and Swollen Behavior of Natural Rubber Filled Groundnut shell, Researcher, Vol. 7, Pp5
7. Blow, C.M. and Hepburn C. (1982): Rubber Technology and manufacture, Butterworth scientific, London 2nd edition 1-540
8. Egwaikhide P.A., Akporhonor E.E., and Okieimen F.E, (2007): Effect of Coconut Fibre Filler on the Cure Characteristics, Physico- mechanical and Swelling Properties of Natural Rubber Vulcanizates, International Journal of Physical Sciences, 2(2), , 39-46.
9. Hepbum, C (1984): Filler Reinforcement of Rubber, Plastics and Rubber International, 9 11.
10. Hussain, A. K., Abdel, K., Ibrahim, A. (2010): Effect of Modified Linen Fibre Waste on Physic-Mechanical Properties and Non-polar Rubber, Journal of Natural Science, Vol. 8, No. 8,, 82-90
11. Ismail H., Rozman H.D., Jaffri R.M, and Ishak Z.A., (1997): Oil Palm Wood Flour Reinforced Epoxidized Natural Rubber Composites: Effects of Filler Content and Size, European Polymer Journal, 33(10-11), 1627-1632.
12. Lovely M., Joseph K.U, and Joseph R., (2006): Swelling Behavior of Isora/Natural Rubber Composites in Oils used in Automobiles, bull. Mater. Sci., 29(1), , 91-99.
13. Marut, A.J., Jekada, J. Z., Okele, I. A., Mohammed, F. and Agho, B. O. (2015): Research and Reviews In. Research and Reviews in Polymers, Trade Science Inc., 6(4), 155–160.
14. Mohanty, A.K., Misra, M., Aizal, L.T. and Kellys (2001): Surface Modification of Natural Fibres and Performance of the Resulting Biocomposites, An Overview of Composite Interfaces, Vol. 8, pp5.
15. Okieimen, F. E. and Akinlabi, A.K. (2002): Processing Characteristics and Physic-Mechanical Properties of Natural Rubber and Liquid Natural Rubber Blends, Journal of Applied Polymer Science, Vol. 85, 1070-1076
16. Okieimen, F.E and Imanah, J.E (2003): Characterization of Agricultural Waste Products as Fillers in Natural Rubber Formulation, Nigeria Journal. Polymer Technology, 3(1), 201-207.
17. Okiemen, F.E and Imanah, J.E. (2005): Physico-Mechanical and Equilibrium Swelling Properties of Natural Rubber Filled with Rubber Seed Shell Carbon. Journal Polymer Material 22 (4): 409.
18. Okwele, A. I., Gimba, C. E., Mamza, P. A. P., and Abba, H. (2018): Hybridisation of Carbon Black: Cashew Nut Shell Powder as Fillers on the Mechanical Properties of Natural Rubber Composites. Composite Materials. Vol. 2, No. 2, Pp. 49-54.
19. Osabohien, E; Adaikpoh, E O; Nwabue, F I (2004): Analysis and Purification of Local Limestone and Poultry Eggshell for $CaCO_3$ Used in Paint Manufacture, Proceedings of the 27th International conference of Chemical Society of Nigeria. Benin-City, Nigeria, 300-304. Publisher Ltd. Lagos, Nigeria, 227-228.
20. Sau K.P., Chaki T.K., and Khastgir D. (1999): Electrical and Mechanical Properties of Conducting Carbon black Filled Composites based on Rubber and Rubber Blends, Journal of Applied Polymer Science, 71, 887-895
21. Sobhy, M.S, Mahdy, M.M, El-Fayoumi, M.A.K, and Abdel-Bary, E.M, (1997): Polymer Testing, 16, 349

22. Sukru, Y.O., Zlem, C, and Ahmet, C. (2008): The Effects of Fibre Contents on the Mechanical Properties of Adobes, Construction volume 38 p 943.
23. Tenebe O.G, Ayo. M.D, Ichetaonye.S.I, Igbonazobi L.C and Eguare K.O, (2013): Effect of Filler Loading on the mechanical Properties of Natural Rubber Filled with Guinea Corn Husk (Holus sorghum) as Filler. International Journal of Life Science, Vol. 2 No. 4, Pp 156-16
24. Tenebe, O. G., Ayo, M. D., Amoke, A., Edegbe, O. C. and Eguare, K. O (2019): Micro-Structural Properties of Natural Rubber Reinforced with Mercerized Rice Husks. Nigerian Research Journal of Chemical Science (NRJCS). Vol. 6, Pp 275-285.
25. Thomas, T., Ayswarya, E.P., and Eby, T.T. (2013): “Nano alumina as reinforcement in natural rubber composites,” International Journal of Innovative Research in Sciences, Engineering and Technology, vol. 21, issue 6, pp. 2319-2370.