International Journal of Physical Sciences Vol. 2 (2), pp. 039-046, February, 2007
Available online at http://www.academicjournals.org/IJPS
ISSN 1992 - 1950 © 2007 Academic Journals

Full Length Research Paper

Effect of coconut fibre filler on the cure characteristics physico–mechanical and swelling properties of natural rubber vulcanisates

Egwaikhide, P. A[1]*; Akporhonor, E. E[2] and Okieimen, F. E[1]

[1]Agricultural Waste Research Laboratory, Department of Chemistry, University of Benin, Benin City
[2]Department of Chemistry, Delta State University, Abraka, Delta State

Accepted 19 January, 2007

Investigation into the effect on the rheological, physico – mechanical and swelling properties of natural rubber vulcanizates using coconut fibre as fillers were carried out. The coconut fibre was characterized in terms of pH, moisture content, ash content, loss on ignition, conductivity, particle size and surface area. The physico – mechanical properties as well as the equilibrium swelling characteristics of the vulcanizates in organic solvents were measured as a function of filler loading and compared with the values obtained using commercial grade carbon black (N330). The coconut fibre showed good processing safety in terms of torques and scorch. For coconut fibre filled vulcanizates, optimum tensile strength of 7.35 MPa at 60 ph was recorded. It was found that vulcanizates with 60 ph showed maximum tensile properties. Hardness of filled vulcanizates with coconut fibre increased with filler loading. Abrasion resistance decreases marginally with increasing filler loading. A flex resistance and percentage compression set decreased with increasing filler loading. The equilibrium sorption in organic solvents of natural rubber vulcanizates filled with coconut fibre and carbon black decreased with increasing filler loading. However, the resistance to swelling of natural rubber compound is dependent on the amount of filler loading: the higher the filler content, the lower the equilibrium sorption values obtained.

Key words: Vulcanizate, rheological properties, abrasion resistance, equilibrium sorption.

INTRODUCTION

The search for means and methods of improving the properties and processing of rubber dates back to over a century ago. One way of achieving this extension of service life of rubber is the incorporation of additives into the polymer matrix.

Additives are materials when incorporated into a polymer base, help to ensure easy processing, reduce cost of product and enhance service properties (Ski, 1970).

The different types of additives used in the processing of rubber into products include, vulcanizing agents accelerator, activator, anti – degradants, fillers, softener, thickners, gel sensitizer, colorant e.t.c. (Okieimen and Imanah, 2003)

Fillers is one of the major additives used in natural rubber compound and has marked effect and influence on rubber materials. Filler functions to modify the physical and, to some extent, the chemical properties of vulcanizate (Drivers, 1979)

The mechanism of reinforcement of elastomers by fillers has been reviewed by several workers (Brennan and Jermyn, 1965). They considered that the effect of filler is to increase the number of chains, which share the load of a broken polymer chain. It is known that in the case of filled vulcanizates, the efficiency of reinforcement depends on a complex interaction of several filler related parameters. There include particle size, particle shape, particle dispersion, surface area, surface reactivity, structure of the filler and the bonding quality between the filler and the rubber matrix.

*Corresponding author. E-mail: pegwaikhide@yahoo.com.

Table 1. Recipe for compounding the natural rubber mixes

Ingredient	(phr) (parts per hundred)
Natural rubber	100
Filler	0 – 80
Zinc oxide	4.0
Stearic acid	2.0
Sulphur	1.5
Mecaptobenzothiazole	1.5
Processing oil	20

In rubber industry, filters that are commonly in use are carbon black, china clay and calcium carbonate. Carbon black is derived from petro-chemical sources but the unstable price of crude oil has led to the search for filler that are derived from other sources (Ski, 1970). Agricu-ltural by-products; maize cobs, cocoa pod husk, sugar cane shaff, rice husk, plantain peel etc. are low cost materials and readily available in large quantity for use everywhere, of which well over 300 million tones are pro-duced annually. In previous reports, the use of cocoa pod husk, rubber seed shells (Okieimen and Imanah, 2003), groundnut husk, plantain peels etc were examined. The results obtained from these studies indicated a potential for the utilization of agricultural residues as fillers in natural rubber compou-nds. In this present study, the effect of coconut fibre on the cure characteristics, physico-mechanical and swelling properties of natural rubber vulcanizate are examined.

MATERIALS AND METHODS

Materials

Crumb natural rubber which conforms to Technically Specified Rubber (TSRIO) but usually (NSRIO) shown as Nigeria Standard Rubber was obtained from 1yayi Rubber Factory, Egba Benin City. The rubber compounding chemicals such as zinc oxide, stearic acid, processing oil, N330 carbon black, sulphur mercaptobenzo-thiozole sulphanamide (MBTS) and tetramethyl thiuram disulphide (TMTD from Vulnay) were of commercial grades. Coconut fibre was obtained from Ihievbe in Edo State, Nigeria.

Filler Preparation

Large quantity of coconut fibre was obtained from the coconut shell. Coconut fibre was dried in an oven at 125℃ to a constant weight. The coconut fibre was ground into fine powder using electric milling machine for corn, cassava and yam flour etc. the ground coconut fibre was therefore sieved with a mesh sieve of size 212 um and retain by a 80 um mesh size.

Compounding

The recipe used in the formulation of the natural rubber compound is given in Table 1. Mixing was carried out on a laboratory two-roll mill in accordance with the method described in the American Society for testing and materials (ASTM – D3184 – 80).

Cure characteristics

The cure characteristics of the compound mixes were measured at 175℃ on an Oscillating Disc Rheometer (ALPHA ODR 2000) in accordance with the 1S0 3417 method.
The cure time (t_{90}), torque (minimum torque (t_{min}), and maximum (t_{max}) were determined from the rheographs. The cure rate and ODR torque were determined using the expression.

$$\text{Cure rate} = \frac{100 \ (S^{-1})}{T_{90} - t_2}$$

$$\text{ODR torque} = \frac{90\ t_{max} - t_{min}}{100} + M_{min}$$

Physico – mechanical properties

Tensile strength

The text specimens were molded in an electrically heated press at conditions predetermined from the rheographs. Tensile properties of the vulcanizates were measured with a Monsanto Tensile Tester (Mode 1/m) at cross – head speed of 500 mm/min using dumb bell test specimens (type H) as contained in ASTMD-412-87 (method A). The tensile strength at break was calculated as the elongation at break.

Compression set

Wallace compression set machine (Mode/Ref No (2, H_2^{50}) was used to determine the compression set of the vulcanizates.

$$\text{Compression set \%} = \frac{t_0 - t_r}{t_0} X \ 100$$

Hardness test

The hardness test of a rubber is the relative resistance of the surface to indentation by an indentor of specified dimension under a specified load. Hardness of the vulcanisates was determined by standard dead load method (BS903 part A 26).

Abrasion resistance

Wallace Akron tester was used in accordance with BS method

$$\text{Abrasive resistance index} = \frac{(S)\ 1000}{T}$$

Swelling properties

The resistance of the vulcanizates to diesel, kerosene and toluene was determined by using the method described in ASTM-D3010. Three different shapes of the cured samples were cut from the 1mm thickness mould and weighed and immersed in air tight containers of diesel, kerosene and toluene solvents at ambient temperature for 24 h. The samples were then removed from the bottles wiped dry with filter paper and weighed immediately. The change in weight of the sample was expressed as percentage swelling.

$$\text{\% Swelling} = \frac{W_2 - W_1}{W_1} x\ 1000$$

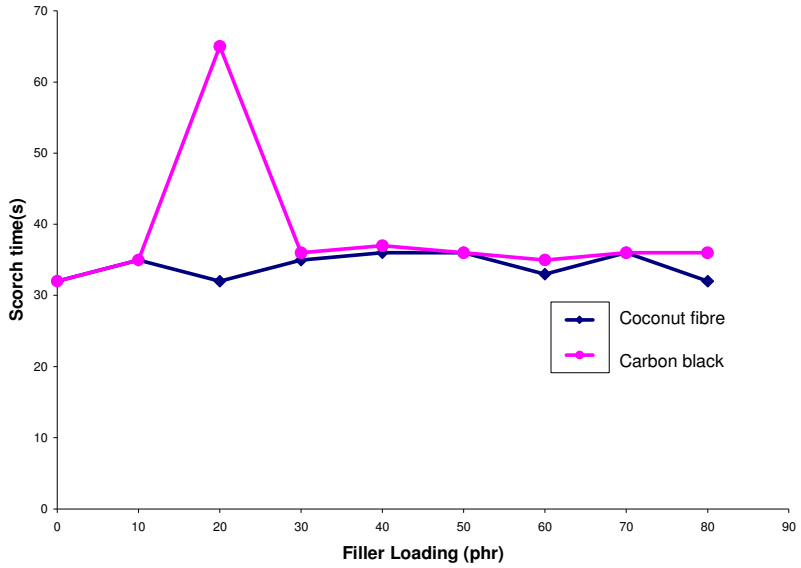

Figure 1. Scorch time – filler loading curve of coconut fibre and carbon black – filled natural rubber vulcanizates

Table 2. Characteristics of coconut fibre

Parameters	Coconut
pH of slurry	6.20
Moisture content (wt%)	10.8
Ash content (%)	2.55
Loss on ignition (875^0C) (%)	37.40
Conductivity (um)	0.82
Length (um)	0.30
Width (um)	0.02
Diameter (um)	0.05
Lumen (um)	0.01
Surface area (cm^3)	0.06

Where W_1 and W_2 are the initial and final weight of the swollen sample respectively.

RESULTS AND DISCUSSION

Characterization of coconut fibre in terms of the moisture content, pH, surface area, ash content and loss on ignition was necessary because the above parameters play an important role in determining the distribution and dispersion of the fillers in natural rubber. Some characteristics of the coconut fibre are given in Table 2.

The physical properties of coconut fibre show that the pH of the slurry is slightly acidic, 6.19. The weight loss on ignition at 875°C is 37.40% for coconut fibre. The weight loss on ignition is a measure of the carbon content lost during combustion and measures the effectiveness of the filter. The higher the values, the greater the reinforcing potential (Okieimen et al., 2003).

Cure characteristic of coconut fibre reinforced natural rubber

Figures 1, 2 and 3 show the cure time; scorch time and maximum torque of the coconut fibre and carbon black filled natural rubber vulcanizates. The difference in cure characteristics may be attributed to the fact that each type of filler possesses different filler properties such as surface area, surface reactivity and particle size (Ishak and Baker, 1995; Hephburn and Blow, 1971). From Figure 1, 2 and 3 the torque values obtained are independent of the filler loading for coconut fibre and carbon black filled vulcanizates but a higher torque value is observed for coconut fibre filled vulcanizates up to 30% filler loading. The torque value of carbon black is observed to be higher at 40% filler loading with increasing filler content. The highest torque value of 9.48 was recorded at 20% filler loading for coconut fibre and the highest torque value of 5.46 was recorded at 60% filler loading for carbon black respectively. The higher maximum torque of 9.48 at 20% filler loading recorded in the case of coconut fibre – filled vulcanizates may be attributed to the nature of the coconut fibre fillers such as surface area, surface reactivity and particle size (Rivin, 1963). In general, a faster cure rate is obtained with fillers having a low surface area, high moisture content and smaller particle size (Wagner, 1976). It was reported that cure rate is directly related to the humidity and water content of the rubber compound. However, in the present study, the most probable factors to account for the obser-ved cure enhancement of carbon black fillers over coconut fibre fillers are surface area, surface reactivity and particle size. It is believed that the smaller the particle size, hence larger surface area, the greater the interaction between

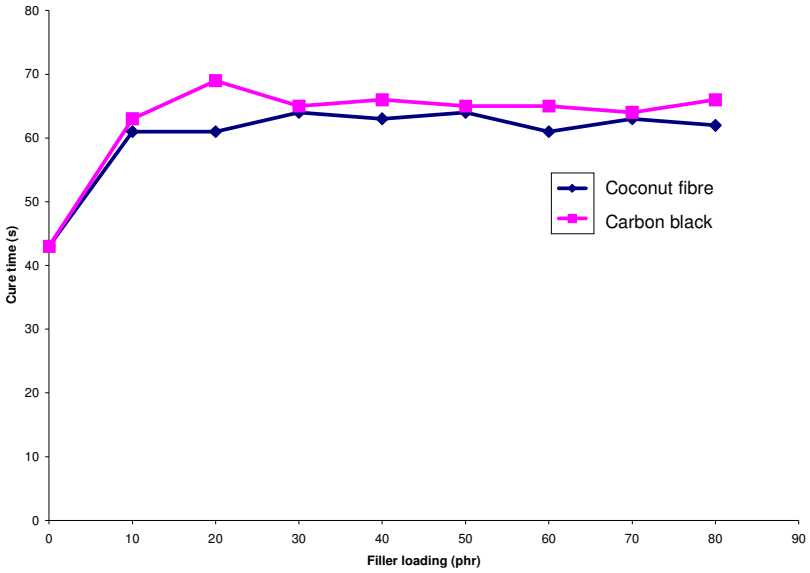

Figure 2. Cure time – filler loading curves of coconut fibre and carbon filled natural rubber vulcanizates.

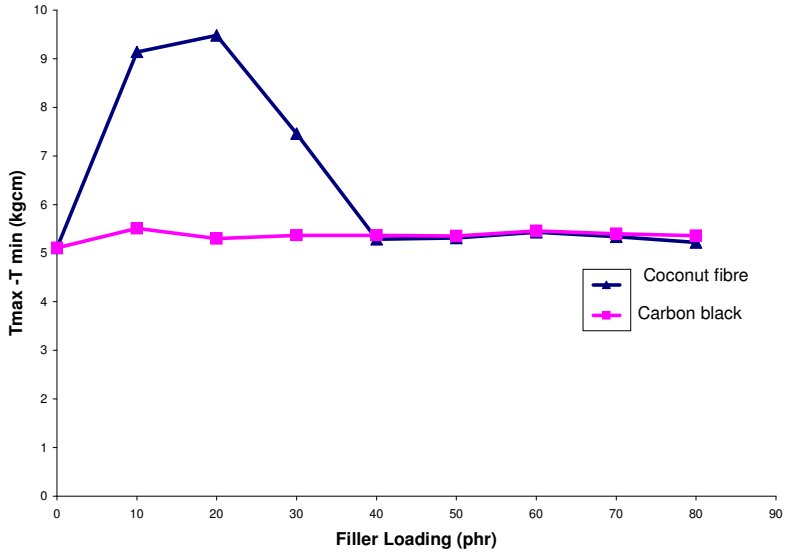

Figure 3. Maximum torque – filler loading curves of coconut fibre and carbon black filled natural rubber vulcanizates.

the filler and rubber matrix.

Mechanical properties of coconut fibre-filled natural rubber vulcanizates Figures 4, 5, 6, 7, 8 and 9 summarizes the physical properties of coconut fibre and carbon black (N330) filled natural rubber vulcanizates. It can be seen that the tensile strength of coconut fibre-filled vulcanizates is inferior to those of commercial fillers. Since coconut fibres have a larger particle size compared to commercial fillers. It has been reported by several workers (Patterman, 1986; Mark, 1964) that significant reinforcement is only attainable when the particle size of the filler is of the order of 0.02 – 0.05 mm. Nasir and Choo (Nasir and Choo, 1989) for instance, found that decreasing the particle size carbon black filler generally enhanced mechanical properties such as tensile strength. A similar observation has been encountered by Parkinson, (1957) in the case of carbon black. The coconut fibre filled – vulcanizate displays lower tensile strength values as compared to the commercial one (carbon black N330) as evident in Figure 4. This may be attributed to the natu-

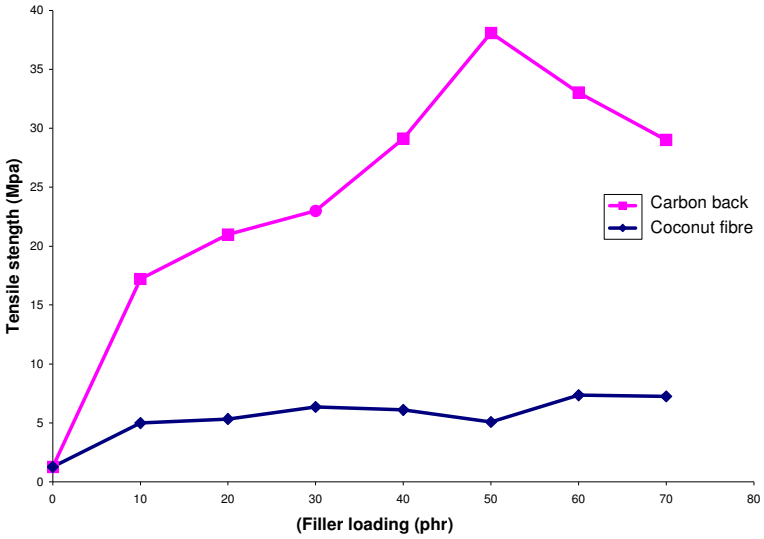

Figure 4. The effect of filler loading on tensile strengths of coconut fibre and carbon black N330.

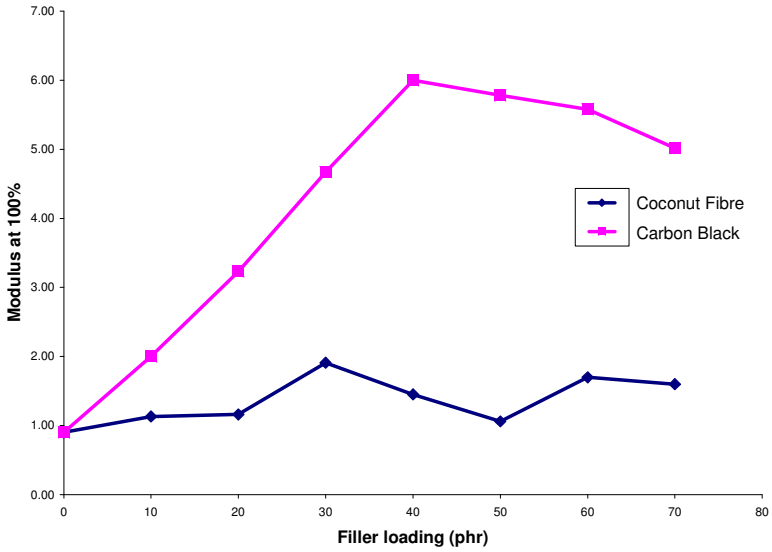

Figure 5. The effect of filler loading on modulus at 100% of coconut fibre and carbon black N330.

re of surface properties, particle size and surface area. Tensile strength is affected by particle size and surface area of the filler. The Tensile Strength increases with increasing particle size of filler (Honday, 1966). In addition, it is also controlled by the nature of both the rubber and filler (Morton, 1987).

As for the modules properties, Figure 5 the trend observed is expected. Coconut fibre –filled vulcanizates has lower moduli then carbon black (N330) – filled vulcanizates. This again may be explained in terms of the difference in the filler properties. Parkinson (1957) and Wagner (1976) reported that the modulus of filled vulcanizates can be enhanced by improving the surface area and surface reactivity of fillers, filler dispersion and filler – rubber interaction. The inferior modulus of coconut fibre – filled vulcanizates may be accounted by two main factors. It is a known fact that those fillers have a larger particle size and hence a smaller surface area than carbon black (N330) fillers. Secondly, coconut fibre fillers show a greater tendency towards filler agglomeration.

The results interpreted by Figure 6, gives a graph of elongation at break as a function of filler loading for both the coconut fibre and carbon black. The values of elongation at break (Eb) decreases with increase in filler loading for

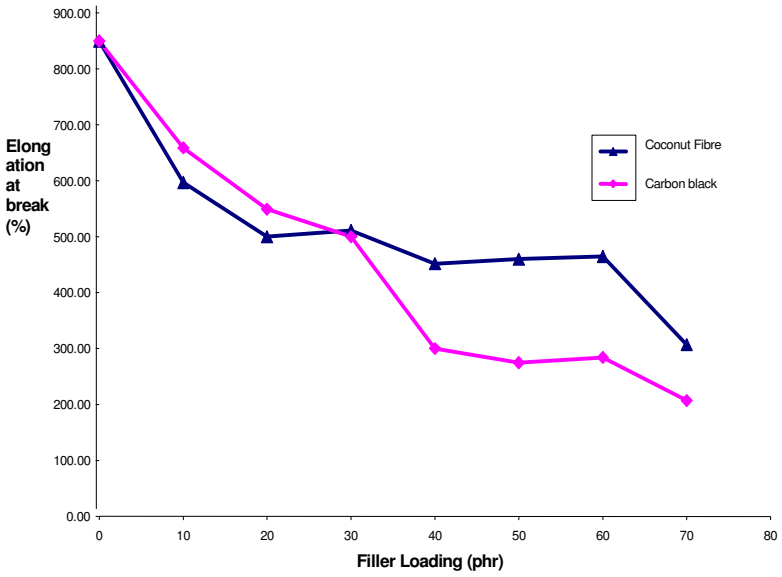

Figure 6. The elongation at break (%) of coconut fibre and N330 – filler vulcanizates.

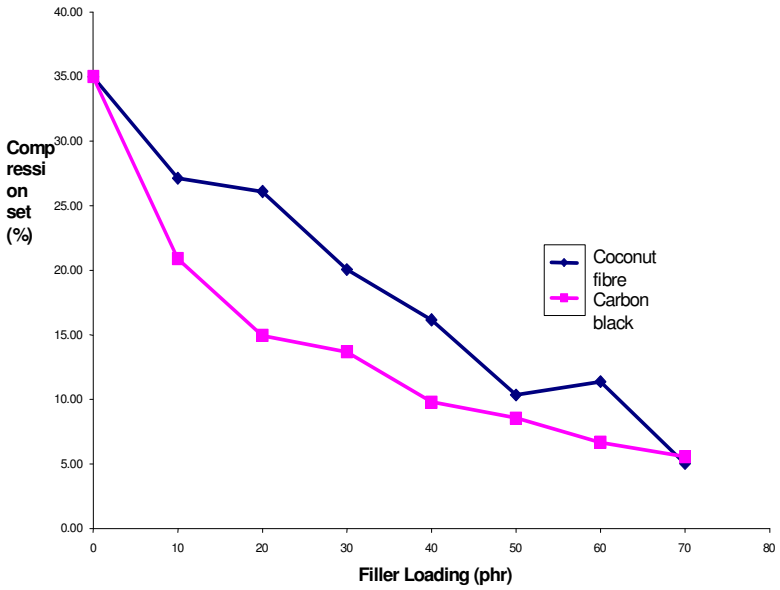

Figure 7. The compression set (%) coconut fibre and N330 – filled vulcanizates.

both the coconut fibre and carbon black vulcanizates. Decreases in elongation at break have 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 (Hephburn and Blow, 1971; Rivin, 1963).

Figure 7, which is the compression, set results showed that unfilled system had the largest compression. Compression decreases with increase in filler loading. The observation may not be unconnected with the amount of filler incorporated into the matrix, the degree of dispersion of the fillers and its particles size. Carbon black has the least compression set.

The hardness Figure 8 of coconut fibre and carbon black vulcanizates increased with increasing filler content. It is expected because as more filler particles get into the rubber, the elasticity of the rubber chain is reduced, resulting in more rigid vulcanizates. The hardness of carbon black vulcanizates is superior to those of agricultural by- products.

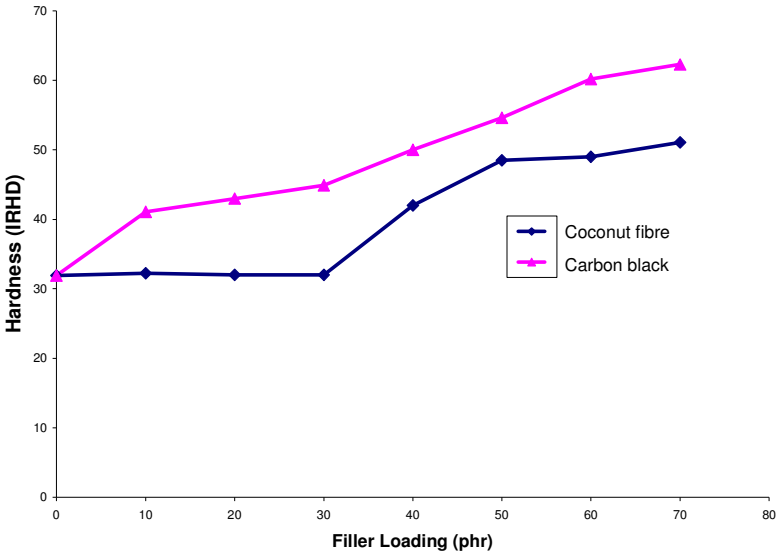

Figure 8. The hardness of coconut fibre and N330 filled vulcanizates

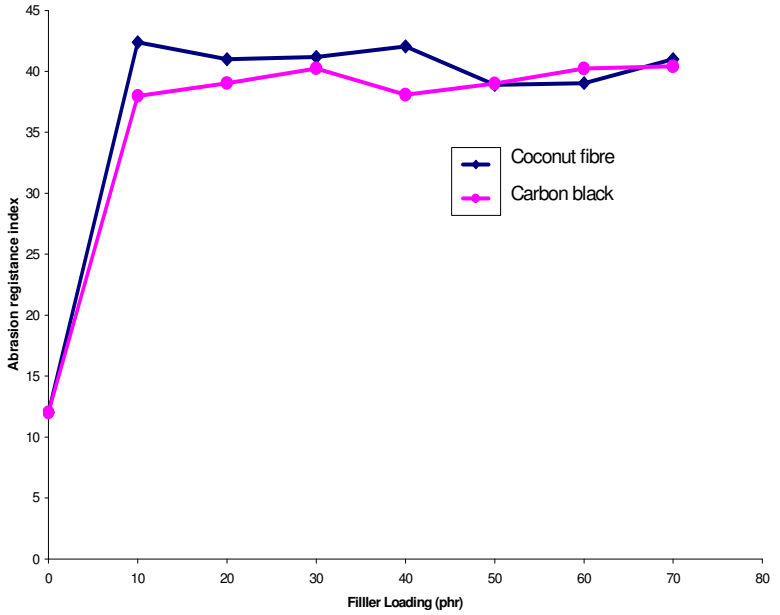

Figure 9. The abrasion resistance of coconut fibre and N330 – filled vulcanizates

The abrasion resistance Figure 9 showed an irregular pattern of increase with increasing filler loading for the filler used. This indicates that filler loading is not a function of the measured parameter. The observation may therefore be attributed to the degree of dispersion of the fillers.

Sorption equilibrium

Table 3 shows equilibrium swelling of coconut fibre filled natural rubber vulcanizates.

Several factors can influence the equilibrium sorption in organic solvent of gum and filled vulcanizates: such factors are level of cross-link, filter dispersion, nature of solvent and fillers. Gent and Lui, (1991) explain why higher sorption values were obtained for kerosene.

Kerosene being a mixture of hydrocarbon C_{10} – C_{14} with a lower molecular weight than diesel C_{14} – C_{19} may be expected to diffuse faster and be accommodated in the rubber matrix with less hindrances. The decrease in sorption with increasing filler loading may be that each filler particle behaves as an obstacle to the diffusing mlo-

Table 3. Equilibrium Sorption in Diesel, Kerosene and Toluene of Natural Rubber Vulcanizate Filled with Coconut Fibre and Carbon Black (N330).

Filler Loading (phr)	Diesel	Kerosene	Toluene
0	280.07	320.37	397.98
10	269.75 (262.17)	330.50 (280.09)	340.00 (266.25)
20	245.07 (247.00)	310.09 (270.09)	340.00 (266.25)
30	219.00 (180.20)	300.02 (259.05)	322.10 (260.00)
40	201.20 (160.75)	270.75 (245.00)	270.75 (255.00)
50	185.09 (145.25)	241.17 (221.27)	262.70 (234.00)
60	139.50 (120.09)	185.17 (200.09)	242.00 (202.07)
70	149.00 (122.00)	160.20 (160.00)	205.33 (190.01)
80	133.20 (111.05)	127.75 (154.00)	192.20 (172.27)

Values with Carbon Black (330) in parentheses

ecule. As filter loading increase in rubber matrix, more and more obstacles are created to the diffusing molecule and thus reduce the amount of penetrated solvent.

Conclusion

The work presented in this paper is preliminary in nature. The effect of natural rubber filler and filler – filler interaction on rubber reinforcement was investigated by the use of coconut fibre as fillers. The main objective or aim of this study is to gauge the possibility of utilizing the low-cost coconut fibre as alternative filler material in natural rubber. The results show that coconut fibre is potential reinforcing filler for natural rubber compounds. However, carbon black (N330) product exhibited relatively better reinforcing properties than coconut fibre. This study indicates that the potential of coconut fibre and other agricultural by-products can be exploited further by controlling particle size and particle distribution, improving filler dispersion and also its surface functionality.

REFERENCES

Hephbum C, Blow CM (1971). Rubber Technology and Manufacture 3rd Edition Butterworth Publishing pp. 188.
Rivin D (1963). Polymer – Filler interaction in rubber reinforcement. Rubb Chem. Tech. 36: 729.
Okieimen FE, Akinlabi AK, Aigbodion AI, Bakare IO, Oladoja NA (2003). Effect of epoxidised natural rubber. Nig. J. Polym. Sci. Technol. 3(1): 233.
Okielmen FE, Imanah JE (2003). Characterization of agricultural waste products as fillers in natural rubber formulation. Nig. J. Polym. Technol. 3(1): 201.
Mark HF (1964). Encyclopedia of Polymer Sciences and Technology, Interscience New York. 12: 42.
Brennan JJ, Jermyn TE (1965). Properties of black reinforced blends of natural rubber and butadiene rubber. J. Appl. Sci. 9: 27–45.
Ski KB (1970). Engineering Material. Properties and selection. Reston Publishing Company. pp. 40.
Honday L (1966). Composite Material. Elsevier. New York. pp. 313.
Morton M (1987). Rubber Technology 3rd Edition Vani Nostrand, New York. pp. 74.
Nasir M, Choo Ch (1989). Chemical modification of natural rubber latex with peracetic acid. Enr. Polym. J. 25: 355.
Wagner MP (1976). Silicone rubber compositions incorporating silicone – treated carbon black 50: 342.
Patterman MQ (1986). Chemorheology of model filled rubber compound during curing. Rubber world 194: 38.
Gent N, Lui GL (1991). Swelling and electrical properties of rubber vulcanizates loaded with paraffin wax. J. Polym. Sci. Polym. Phys. 29: 1313–1319.
Parkinson D (1957). Reinforcement of Rubbers. Lakeman and Co. London. pp. 12.
Donelly PJ (1999). Medium thermal black for cost reduction and property enhancement. Rubb. Technol. Int. pp. 52–56.
Drivers WE (1979). Plastic Chemistry and Technology. Van Nostreand Company, London pp. 243.
Ishak ZAM, Baker AA (1995). An investigation on the potential of rice husk ash as fillers for expoxidised natural rubber. Enr Polym. J. 31(3): 259.