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# The Characterization of Carbonised Coconut Fibre as Fillers in Natural Rubber Formulations

[1]A.P. Egwaikhide, [2]E.E. Akporhonor and [1]F.E. Okieimen
[1]Department of Chemistry and Center for Biomaterial Research, University of Benin, Nigeria
[2]Department of Chemistry, Delta State University, Abraka, Nigeria

**Abstract:** Carbonized coconut fiber of approximate particle sizes 100 and 50 μm were characterized in terms of pH, loss on ignition, surface area, aggregate structure and moisture content, used as filler in compounding natural rubber (NSR10) and vulcanized using efficient vulcanization system and compared with a commercial filler (N330). The cure characteristics of the various mix were determined at 185°C using Monsanto rheometer model ODR2000. The Mooney viscosity was carried out using MK111 Mooney viscometer. Tensile strength, modulus, elongation at break, compression, hardness and abrasion resistance were measured as a function of filler volume fraction loading. It was found that vulcanizates with 0.36 filler volume fraction showed optimum tensile strength of 38 mpa for carbon black and 0.43 filler volume fraction of 10 mpa for carbonized coconut fiber. Hardness property increased with increased filler loading for both fillers and carbon black filled vulcanizates showing superior hardness.

**Key words:** Carbonization, coconut fiber, natural rubber, filler volume fraction, commercial filler, reinforcing filler, vulcanization

## INTRODUCTION

In the rubber industry, compounding and vulcanization transform raw rubber into a range of materials suitable for application in various uses and in different service environment (Hofman, 1967). Natural Rubber (NR) alone do not possess the necessary tensile, modulus and hardness that is required by rubber manufacturers. It is therefore necessary to incorporate materials which will increase the above characteristics to the desired level (Studebaker, 1957). Among some of the materials used in natural rubber compounding are fillers. These fillers assist in attaining the desired characteristics and constitute the second largest material in term of quantity in a rubber compound after the rubber itself (Brennan and Jermyn, 1965). Particulate fillers such as carbon black, calcium carbonate and China clay are essentially and widely used as reinforcing filler in the industries. Calcium carbonate ($CaCO_3$) has attracted considerable interest in recent years due to its availability and low cost and the dependence of the traditional carbon black on crude oil (Danneberg, 1981).

One of the mechanism by which particulate fillers reinforces elastomers is that reported by Flemimert (1957). They considered that the effect of filler is to increase the number of chains, which shared the load of a broken polymer chain.

Agricultural residues are low-cost materials and readily available in large quantity for use everywhere (Imanah and Okieimen, 2003; Hess and Medelia, 1978). In a previous report, the utilization of cocoa pod husk rubber seed shells etc. as fillers in natural rubber was examined (Odian, 1981; Okieimen and Imanah, 2003; Egwaikhide *et al*., 2007). As for coconut fibre not much has been reported about its application. This study present results of assessment and utilization of carbonized coconut fibre as filler for the production of rubber products.

**Corresponding Author:** A.P. Egwaikhide, Department of Chemistry and Center for Biomaterial Research, University of Benin, Nigeria

## MATERIALS AND METHODS

### Materials

The coconut fibre used in this study were obtained from Ihievbe market, Nigeria and air-dried, ground and sieved through a mesh size of 80 μm. The sieved coconut fibre powder was carbonized at 200°C using the procedure described by Ishak and Baker (1995). Natural rubber (NSR10) was purchased from Iyayi Rubber Factory Egba, Benin City Nigeria and was used as received. Carbon black and the other rubber additives such as stearic acid, MBTS, processing oil, sulphur, zinc oxide used in the compounding were of industrial grade and all materials were used as supplied.

### Characterization of Coconut Fibre

The chemical and physical properties of carbonized coconut fibre and carbon black are given in Table 1. The carbonized coconut fibre powders were characterized in terms of loss on ignition (ASTM 1509 1983), moisture content, pH of slurry (ASTM 1512, 1983) surface area (ASTM 1510, 1983) and aggregate structure (Hepburn, 1984).

Moisture content was determined by weighing 5 g of dried coconut fibre powder into petri dish. The sample was placed in an oven for 1 hour at a temperature of 125±1°C and left to cool down in a desiccator. The change in weight was calculated as a percentage of the sample weight (ASTM 776, 1983). The pH of the powder slurry was determined with a pH meter. Weight loss on ignition, was determined by weighing 2 g of dried carbonized coconut fibre in a 30 $cm^3$ porcelain crucible which has been placed in a electric furnace at a temperature of $825^{+}$- 25°C for 4½ h. Weight loss on ignition was calculated based on the amount of solid residue.

### Compounding

Simple mix formulations as shown in Table 1 were used with batch factor of 4. An efficient vulcanization system was chosen. The elastomer was placed in a water-cooled laboratory size (160×320 mm) two-roll mill. The mill was made to attain a maximum temperature of about 80°C to prevent the compound from scorching.

### Processing Characteristics

A Monsanto rheometer model ALPHA ODR 2000 was used to determine the processing characteristics of the compound mixes. The discs were set to an arc angle of 0.50°C at a curing temperature of 185°C. The scorch time, cure time and torques of filled and unfilled natural rubber, were determined from the rheometer rheograh as shown in Table 2 (Yehia and Stoll, 1987).

### Mooney Viscosity

The Mooney Viscosity of the mixes was carried out using Negretti MK 111 Mooney Viscometer. There is preheating period of 1 min at 100°C. Rotor was there after engaged to run for 4 min. Mooney Viscosity displaced automatically and it is expressed as ML (1+4) 100°C.

Table 1: Formulations for CCF and CB filled natural rubber

| Materials (phr) | |
|---|---|
| Natural rubber NSR 10 | 100.00 |
| Stearic acid | 4.00 |
| Filler | 0.70 |
| Zinc oxide | 2.00 |
| MBTS | 2.00 |
| Processing oil | 2.00 |
| Sulphur | 4.50 |

Table 2: Cure characteristics of NR filled CB and CCF

| Filler volume fractions | Time (sec) $Ts_1$ | $Ts_2$ | $Ts_{10}$ | $Ts_{50}$ | $T_{90}$ | Min torque ($M_L$) | Max torque ($M_H$) | ODR |
|---|---|---|---|---|---|---|---|---|
| 0 | 0.39 | 0.46 | 0.33 | 0.60 | 1.84 | 1.37 | 6.90 | 6.24 |
| 0.07 | (0.28) | (0.35) | (0.25) | (0.39) | (0.63) | (1.39) | (6.90) | (6.35) |
| | 0.38 | 0.43 | 0.33 | 0.61 | 1.82 | 1.72 | 11.81 | 9.09 |
| 0.14 | (0.38) | (065) | (0.17) | (0.38) | (0.69) | (1.35) | (6.65) | (6.12) |
| | 0.38 | 0.45 | 0.33 | 0.61 | 1.82 | 1.77 | 12.18 | 9.39 |
| 0.29 | (0.29) | (0.36) | (0.25) | (0.40) | (0.65) | (1.41) | (6.78) | (6.24) |
| | 0.39 | 0.46 | 0.35 | 0.60 | 1.86 | 1.80 | 8.20 | 5.78 |
| 0.36 | (0.30) | (0.37) | (0.25) | (0.41) | (0.66) | (1.45) | (6.87) | (6.33) |
| | 0.39 | 0.46 | 0.33 | 0.60 | 1.83 | 1.84 | 7.96 | 5.53 |
| 0.43 | (0.29) | (0.36) | (0.25) | (0.41) | (0.65) | (1.50) | (6.65) | (6.32) |
| | 0.39 | 0.43 | 0.30 | 0.58 | 1.81 | 1.74 | 7.92 | 5.55 |
| 0.50 | (0.28) | (0.35) | (0.24) | (0.40) | (0.65) | (1.48) | (6.94) | (6.39) |
| | 0.36 | 0.46 | 0.34 | 0.60 | 1.84 | 1.71 | 5.58 | 5.58 |

Cure characteristics of natural rubber filler with carbon black in parentheses

## Measurement of Physico-Mechanical Properties

### Tensile Strength

Tensile properties of the vulcanizates were measured with a Monsanto tensile Tester (model$^1/_m$) at a cross-head speed of 500 mm min$^{-1}$ using dumb bell test specimens (type 1) as contained in ASTMD-412-87 method A.

### Compression Set

Procedure adopted for the determination of compression set was based on ASTM 385 (1983e). Wallace compression set machine model with serial No C84025/2 was used (BS 903, 1986).

### Hardness Test

This test was carried out using the Wallace hardness tester of model C8007/25 in accordance with ASTM 1415 (1986) method.

### Abrasion Resistance

The abrasion Resistance was based on DIN to 150 4669 (1948) Akron to BS 903 (1986), part 49 method C using Croydon Akron-Wallace Abrader serial No. C84025/1.

## RESULTS AND DISCUSSION

The physico-chemical properties of carbonized coconut fibre and that of carbon black (N330) (Table 3). Results of fillers used (Table 3) showed that weight loss on ignition at 825°C, are 76.40 (CCF) and 92.8 (CB), respectively. Weight loss on ignition is a measure of the carbon content lost during combustion and it measure the effectiveness of the filler. The higher the values of weight loss on ignition, the greater the reinforcing potential. The values obtained, indicates that carbon black is more reinforcing than carbonized coconut fibre.

The iodine adsorption number provides an estimate of the filler surface area. The results (Table 3) indicate that surface area of carbonized coconut fibre is lower than surface area of carbon black (N330). The larger the value of the surface area, the more reinforcing the filler.

### Mooney Viscosity

The dependence of Mooney Viscosity measured at 100°C on filler loading (Fig. 1, Table 4). The plot of relative Mooney Viscosity, $ML^R$ was used to illustrate the effect of the degree of filler loading.

Table 3: Chemical and physical properties of carbonized coconut fibre (CCF) and CB (N330)

| Parameters | CCF | CB |
|---|---|---|
| Moisture content (125°C) | 6.80 | 2.40 |
| pH | 6.35 | 6.50 |
| Loss on ignition (825°C) | 76.40 | 92.80 |
| Surface area | 62.06 | 81.42 |

Table 4: Mooney Viscosity of filled natural rubber

| Volume fraction of filler | 0 | 0.07 | 0.14 | 0.29 | 0.39 | 0.43 | 0.55 |
|---|---|---|---|---|---|---|---|
| Mooney Viscosity of CCF | 32 | 33 | 37 | 40 | 44 | 49 | 53 |
| Mooney viscosity of CB | 32 | 35 | 41 | 48 | 63 | 76 | 77 |

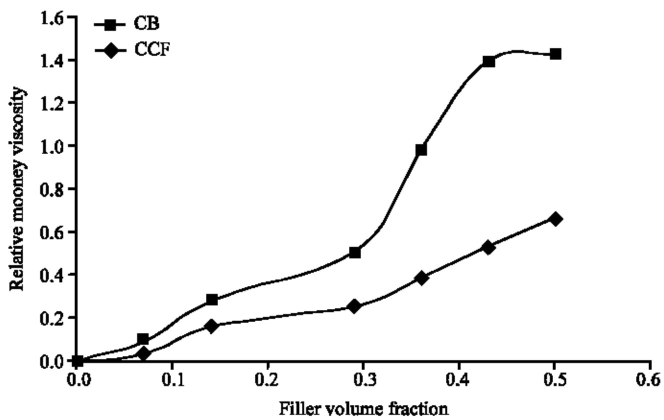


Fig. 1: Relative mooney viscosity as a function of volume fraction for CCF and CB

$$ML^R = \frac{ML_F}{ML_O} - 1$$

Where:
$ML_F$ = The Mooney viscosity of the filled compound
$ML_O$ = The Mooney viscosity of the gum

For both fillers, the relative viscosity increases with filler loading. At small concentrations, there is no significant difference in $ML^R$ between the two fillers. However at high loadings, considerable increases in relative viscosity are observed for carbon black. This is due mainly to the agglomeration of carbon black aggregated. Abrupt rise in $ML^R$ indicates that agglomeration is so strong that it cannot be dispersed during shearing. The higher the value of Mooney Viscosity, the better for the material. It gives an ideal about the crosslink density (Wolff and Wagner, 1972).

**Processing Characteristics**

Analysis of the Monsanto reheograph facilitates the determination of the various cure related parameters.

ODR is the total energy or force required to cure the rubber (Fig. 2). Low ODR is advisable. The maximum torque increases with increasing percentage filler loading for both until fillers a maximum is attain at 12.18 (CCF) before a decline. The optimum cure and scorch time of carbonized coconut fibre exhibit a retardation in cure rates with the torques at maximum, lower for carbonized coconut fibre than carbon black. This trend in cure characteristics may be attributed to differences in the filler properties.

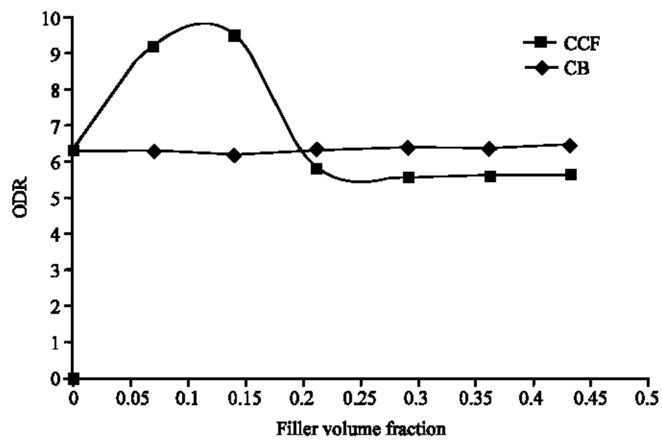


Fig. 2: ODR as a function of filler loading for CCF and CB

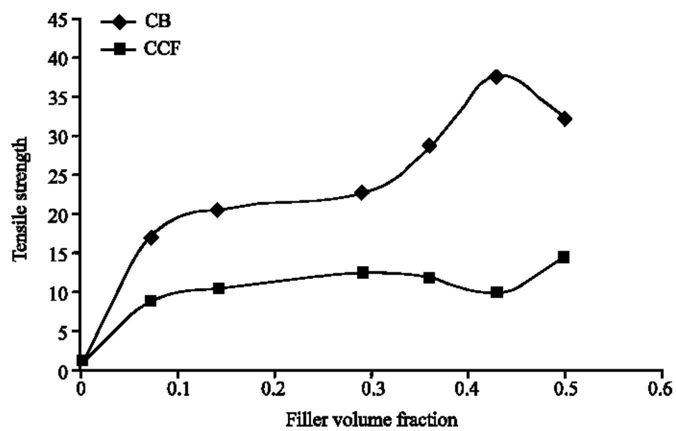


Fig. 3: Tensile strength as a function of volume fraction for carbonized coconut fibre and carbon black (N330)

The cure enhancement in the case of carbonized coconut fibre over carbon black-filled vulcanizates can be associated with the filler related parameters such as surface area, surface reactivity, particle size and moisture content. It is known that alkalinity accelerates cure rate while acidity retards it (Wagner, 1976). It has also been reported that cure rate is directly related to the humidity and water content of the compound mix (Butler and Freakley, 1991). In general, faster cure rate is obtained with fillers having high moisture content and more acidic. However in the present study, the most probable factors to account for the observed cure enhancement of carbonized coconut fiber over carbon black are moisture content and pH. It is believed that the smaller the particle size, hence larger surface area, the greater the interaction between the filler and rubber matrix. Thus a higher restriction to molecular motion of the macromolecules is expected. In other words, the addition of fillers of a smaller size tends to impose extra resistance to flow.

**Physico-Mechanical Properties of Vulcanizates**

In both carbonized coconut fiber and carbon black-filled systems, the tensile strength increased with increasing filler content until a maximum level is reached at 14.70 mpa for carbonized coconut fiber and 38.09 mpa for carbon black respectively (Fig. 3).

A further increase in filler loadings reduces the respective properties. As the filler loading is increased, eventually a level is reached, where the filler properties or aggregates are no longer adequately separated or wetted by rubber phase. This reduction in tensile and tear strength may be due to agglomeration of the filler particles to form a domain that acts like a foreign body. Carbon blacks exhibit a higher tensile strength at any particular loading of filler. The superior tensile strength of CB over CCF may also be due to better filler dispersion, surface reactivity and bonding quality between the filler and rubber matrix.

Modulus (Fig. 4) at 100% for carbon black filled vulcanizates is higher than that of carbonized coconut fibre filled vulcanizate. In reinforced systems, strong carbon polymer bonds are formed.

Elongation at break is a function of filler loading for both fillers (Fig. 5).

The values of Elongation at break (Eb) decreases with increase in filler content of the mix for both fillers. Decrease in elongation at break with increasing filler loading has been explained in terms of adherence of the polymer phase leading to the stiffening of the polymer chain and hence resistance to stretch when strain is applied (Vin, 1963).

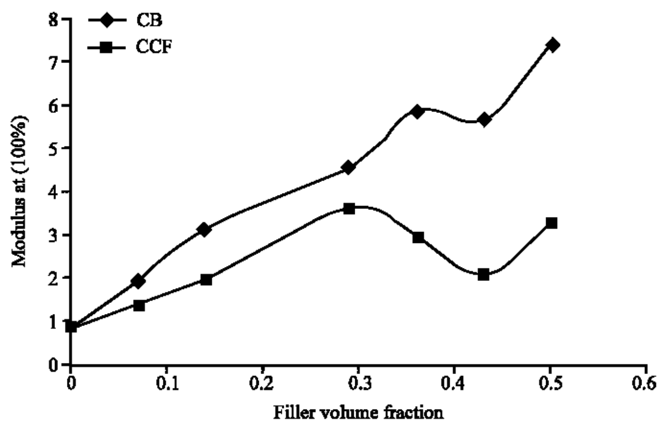


Fig. 4: Modulus at 100% as a function of volume fraction for carbonized coconut fibre and carbon black (N330)

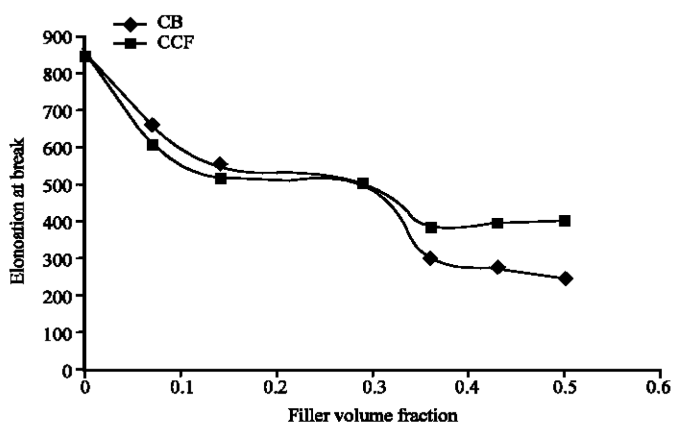


Fig. 5: Elongation at break (%) as a function of volume fraction for carbonized coconut fibre and carbon black (N330)

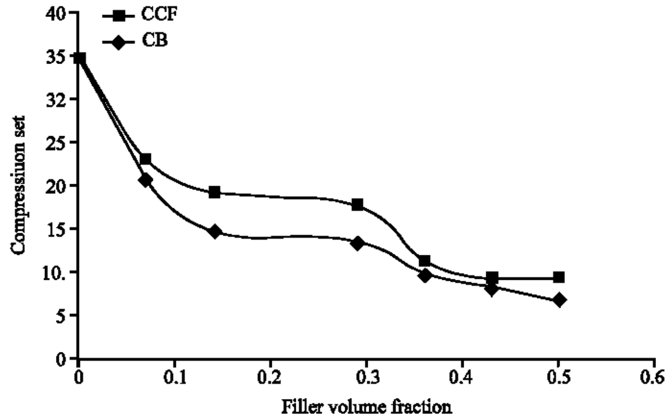


Fig. 6: Compression set as a function of volume fraction for carbonized coconut fibre and carbon black (N330)

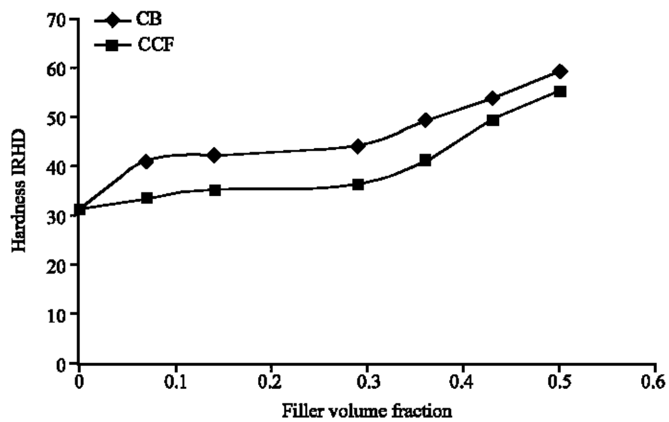


Fig. 7: Hardness IRHD as a function of volume fraction for carbonized coconut fibre and carbon black (N330)

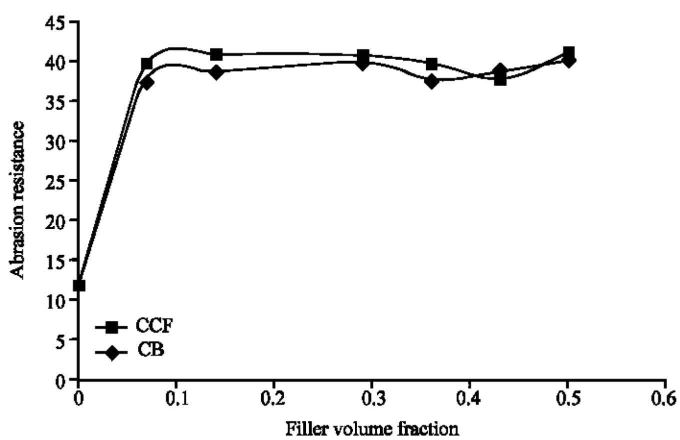


Fig. 8: Abrasion resistance as a function of volume fraction for carbonized coconut fibre and carbon black (N330)

In the case of compression set (Fig. 6) the curve showed that unfilled stock has the highest compressibility of 35.00%. As filler loading increases for the filled stock, the % compression decreases.

Compressibility therefore could be dependent on the amount of filler incorporated into the rubber.

The hardness for both CB and CCF-filled vulcanizates increased with increasing filler content (Fig. 7).

As more filler particles get into the rubber, the elasticity of the rubber chain is reduced, resulting in more rigid vulcanizates. Vulcanizates filled with carbon black showed more improvement in degree of hardness than those filled with carbonized coconut fibre.

Abrasion resistance (Fig. 8) showed and irregular pattern of increase with increasing filler loading for both fillers. This indicates that abrasion resistance is not a function of filler loading and it is due to the degree of fillers dispersion.

## REFERENCES

150, 4669, 1948. Testing abrasion resistance of cure compound mixes.

ASTM 1509, 1983. Standard method of testing hear loss.

ASTM 1512, 1983. Standard method for testing pH value.

ASTM 1510, 1983. Method for testing iodine adsorption number.

ASTM 385, 1983. Method for testing compression set of rubber.

ASTM 776, 1983. American society for testing and material.

Brennan, J.J. and T.E. Jermyn, 1965. J. Applied Sci., 9: 27-49.

BS 903, 1986. Part A26 standard method of rubber hardness.

Butler, J. and P.K. Freakley, 1991. Rubb. Chem. Technol., 65: 374.

Danneberg, E.M., 1981. Proceeding of International Rubber Conference, Lougborough.

Egwaikhide, P.A., E.E. Akporhonor and F.E. Okieimen, 2007. Effect of coconut fibre filler on the cure characteristics, physico-mechanical and swelling properties of nr vulcanizates. Int. J. Phys. Sci., 2: 39-46.

Flemimert, G., 1957. Light reinforcement filler. A Paper Presented Before the Swedish Institute of Rubber Technology.

Hepburn, C., 1984. Filler reinforcement of rubber. Plastics and rubber international No. 9, pp: 11-15.

Hess, G.A.M. and W.M. Medelia, 1978. Reinforcement of elastomer with carbon black in plastic and Rubber Processing, 1.

Hofman, W., 1967. Vulcanization and Vulcanizing Agents. MacLaren and Sons Ltd., London, pp: 300.

Imanah, J.E. and F.E. Okieimen, 2003. Rheological and mechanical properties of nr reinforced with agricultural by product. J. Applied Polym. Sci., 90: 3917-3722.

Ishak, Z.A. and A.A. Baker, 1995. An Investigation on the potential of rice husk ash as fillers for Expoxdised Natural Rubber (ENR) Eur. Polym. J., 31: 259.

Odian, G., 1981. Principles of Polymerisation. 2nd Edn., Wiley-Inter-Science, New York, pp: 128.

Okieimen, F.E. and J.E. Imanah, 2003. Characterisation of Agricultural waste Products as Fillers I Natural Rubber Formulation. Nig. J. Polym. Sci. Technol., pp:

Studebaker, M., 1957. Rubb. Chem. And Technol., 30: 1400.

Vin, D.R., 1963. Rubb. Chem. Technol., 36: 729.

Wagner, M.R., 1976. Rubb. Chem. Technol., 49: 704.

Wolff, S. and M.J. Wagner, 1972. The effect of the surface energies of fillers on elastomer reinforcement. Rubb. Chem. Technol., 65 : 329-342.

Yehia, A.A. and B. Stoll, 1987. Kauts. Gummi kunstz, pp: 40.