EVALUATION OF PHYSICO-MECHANICAL PROPERTIES OF SPONGE-GOURD FIBRE REINFORCED NATURAL RUBBER COMPOSITES

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CERTIFICATION

This is to certify that this research work entitled **"EVALUATION OF PHYSICO-MECHANICAL PROPERTIES OF SPONGE-GOURD FIBRE REINFORCED NATURAL RUBBER COMPOSITES"** was carried out by **EGUAVOEN, ENDURANCE OROBOSA (20134869598)** in partial fulfillment for the award of the Degree of Master of Science (M.Sc) in Polymer Science and Engineering, in the Department of Polymer and Textile Engineering, Federal University of Technology, Owerri, Imo State, Nigeria, under my supervision.

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ABSTRACT

This study involved the separate use of carbonized and uncarbonized sponge-gourd filler to reinforce natural rubber (NR). Carbon black (CB) was also used as well. The fillers were all sieved through a 150μm mesh. Carbonized and uncarbonized sponge-gourd were characterized in terms of moisture content, loss on ignition, density, pH and iodine adsorption number before use. The fillers were used in compounding NR at varying filler loading (3phr, 5phr, 10phr, 15phr and 20phr). The composites were prepared using a two-roll mill. The specimens were moulded using a compression moulding machine. The mechanical properties of the various composites samples prepared were then examined. The results of the tests obtained for the NR- CS composites were compared with those of NR-US and also with those of NR-CB composites. Though an irregular increase was observed, tensile strength result showed that composites of CS and US fillers started increasing at 10phr whereas those of CB started at 5phr. The maximum tensile strength for CB and CS composites were obtained at a filler loading of 15phr. For modulus, an irregular increase was observed, with CB having the highest and was followed by CS. The result for hardness showed a general increase with increase in filler loading. For elongation and compression set, the result showed a decreasing trend for all the fillers to a comparable level with CB. For abrasion resistance, an irregular trend was observed. In all, it was observed that carbonized sponge-gourd presented better potentials for reinforcement than uncarbonized sponge-gourd.

Keywords: Sponge-gourd, carbonization, characterization, natural rubber and fillers.

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CHAPTER ONE

INTRODUCTION

1.1 Background of Study

Rubbers are organic polymer materials which can be stretched to about 200% of its original length and snap back to the original length immediately the deforming force is removed (Aguele et al, 2014).

It is important since it is renewable, inexpensive, creates no health hazard problems, possesses high strength and modulus due to strain-induced crystallization, shows superior building tack essential in products like hoses, belts, tyres etc. and possesses good crack propagation resistance. Nevertheless, NR bears certain disadvantages in that its strength is lower than that of other traditional materials. Thus the products based on NR often suffer from tearing, breakage, puncture and creep. Besides, NR possesses poor crack initiation resistance, ozone and oil resistance. One way of achieving the extensions of service life of rubber products is the incorporation of additives into the polymer matrix (Tenebe et al, 2013). Fillers represent one of the most important additives used in rubber compounding. Carbon black is always considered the most commonly consumed reinforcing filler in the rubber industry (Onyeagoro, 2012). However, their use has certain disadvantages: dark colour, contamination, pollution and non-renewable petroleum origin. These disadvantages coupled with strong environmental regulations worldwide have resulted in a shift to alternative sources like natural fibres. Himadri and Dipul (2014) foresaw benefits of using natural fibres: lower levels of skin and respiratory system irritation during handling; reducing tool wear during processing; good recyclability; abundant supply; low cost; low density; high specific strength-to- weight ratio; non-toxicity and biodegradability.

1.2 Statement of Problem

Carbon black is always considered the most effective reinforcing filler in the rubber industry. This is because of the excellent properties it gives to the rubber compound (Tsubokawa et al, 1993). However, in spite of its unique properties and applications, intensive research is ongoing in search for a suitable alternative in the rubber industry. This is not unconnected with the fact that it is a product of petroleum and therefore non-renewable material. This is in addition to other associated problems such as dark colour and contamination as well as high cost. Besides, the International Agency for Research on Cancer (IARC) evaluation is that, "carbon black is possibly carcinogenic to humans (Group 2B)". Short-term exposure to high concentrations of carbon black may produce discomfort to the upper respiratory tract, through mechanical irritation. These identified draw backs associated with the use of carbon black necessitated the drive by researchers for suitable alternatives to carbon black. Sponge-gourd, the fruit of luffa cylindrica (luffa aegyptiaca) has caught the attention of researchers considering its outstanding properties like high specific strength and modulus, low density, renewability and no health risk. An important factor to consider in the choice of a natural fibre in composite application is availability. Luffa cylindrical meets these criteria since it is in abundance in many parts of Nigeria.

1.3 Objectives of Study

The main objective of this study is to evaluate the physico-mechanical properties of sponge-gourd fibre reinforced natural rubber composites. The specific objectives are to:

(i) Characterize the physical properties of sponge-gourd

(ii) Evaluate critically, the effect of filler loading on the mechanical properties of natural rubber filled carbon black, carbonized and uncarbonized sponge-gourd composites.

(iii) Comparatively study the effect of carbon black, carbonized and uncarbonized sponge-gourd filler on the mechanical properties of NR composites.

1.4 Justification of Study

The intense search for renewable fillers as alternative to carbon black is indeed an indication that we have approached the limit of non renewable materials in our environment. It is interesting to note that a recent survey of literature shows a significant increase in the number of articles relating to use of sponge-gourd in the polymer industry. For instance, it has been used in the reinforcement of polyester resins, epoxy resins, resorcinol formaldehyde etc. But unfortunately, the survey reveals that no attempt has been made to utilize this unique material in the reinforcement of natural rubber, even though natural rubber has been reinforced with other natural fibres like coconut coir, bamboo fibre, dikanut shell etc. The importance of research in this field becomes even more apparent when other researchers mourn the inadequate research in this area. Paglicawan et al. (2005) complained of no study on the properties of luffa fibre as reinforcement material for polymer-bonded composites, Tanobe et al. (2004) decried a situation where some fibres have been extensively investigated and used in composites and medical purposes, many other less known fibres find limited applications as a result of lack of available scientific data regarding their structure and properties and finally Jianhu et al. (2012) lamented that sponge-gourd have not yet had their potentialities fully explored. Thus, this research work is crucial, as it is an attempt to advance the advantage inherent in the use of sponge-gourd as a reinforcement material in NR composites.

1.5 Scope of Study

This study is limited to the following areas:

- Characterization of the physical properties of sponge-gourd,
- Determination of the effect of filler loading on the mechanical properties of NR composites,
- Comparative study of the effect of carbon black, carbonized and uncarbonized sponge-gourd filler on the mechanical properties of NR composites.

CHAPTER TWO

LITERATURE REVIEW

2.1 Natural Rubber

There are different types of rubber including natural rubber (NR) and a variety of synthetic rubbers. NR is obtained from rubber tree (Hevea brasiliensis) in the form of field latex (Katchy, 2008). Among various rubbers, natural rubber (cis-1,4-polyisoprene) is very important because it possesses the general features of other rubbers in addition to the following characteristics. NR is produced by plants so it is renewable, inexpensive and creates no health hazard problems. It possesses high tensile strength and modulus due to strain-induced crystallization. It possesses good crack propagation resistance.

There are various ways of grading dry rubber. These are visual methods, technical specifications and grading as per specification of the buyer. Visual aspects are made use of in conventional grading. The darker the rubber, the lower the grade due to more dirt content. Technical grading is done according to the composition of the rubber, source material and initial plasticity. The main criterion is the dirt content. Table 2.1 gives the NSR-10 specifications for natural rubber.

Table 2.1: Characteristics of Nigerian Standard Rubber (NSR) grade 10 (Osabohien et al, 2015).

Parameters	Values
Dirt content (%)	0.03
Ash content (%)	0.20
Volatile matter	0.41
Nitrogen content	0.25
Plasticity retention index (PRI)	70.86
Mooney viscosity ML (1+4) 100℃	85.00

2.2 Composites

It is a truism to say that only a few polymers are used in their pure form. Thus according to Elias and Vonwinkel (1986), majority of them are combined with small quantity of some other materials called compounding ingredients. In addition to these ingredients, a variety of particulate or fibrous materials are mixed with polymers resulting in heterogeneous, multiphase system. According to Aguele et al. (2014), composite can be generally defined as a combination of two or more components differing in form or composition on a macroscale, having recognizable interfaces between its distinct phases. The different phases are combined carefully to achieve a system with more useful structural or functional properties non-attainable by any of the constituents alone. In addition to having a distinct interface, two other criteria have to be satisfied by a composite material. In the first place, both the constituents are to be present in reasonable proportions, i.e. greater than 5%. Lastly, the constituent phases should possess different properties so that properties of the composite are noticeably different from the properties of both the constituents.

Composites can be mainly classified on the basis of matrix and type of reinforcement. The classifications according to matrix type are ceramic matrix composites (CMC), metal matrix composites (MMC) and polymer matrix composites (PMC). The classifications according to type of reinforcement are particulate composites (composed of particles), fibrous composites (composed of fibres) and laminate composites (composed of laminates). Fibrous composites can be further sub-divided on the basis of natural or synthetic fibre. In fibrous composites, fibres are the load carrying members, while the surrounding matrix keeps them in desired location and orientation. The matrix acts as a load transfer medium, provides shape to the composite structure and protects the fibres from environmental damage. It also gives toughness and compressional strength to the composite. Certain factors are essential in the selection of suitable fibres. They are the required value of stiffness and tensile strength of the composite, thermal stability, adhesion of fibres and matrix, dynamic and long term behaviour, price and finally processing costs.

Besides, composites can be classified on the basis of other criteria like the reinforcing nature of filler, shape of filler and distribution of filler. Fillers can be classified depending on whether it is reinforcing or non-reinforcing. The reinforcing filler can be divided into fibrous or particulate structures. The shape of the reinforcing particles can be spherical, cubic or any other regular or irregular geometry. For particulate reinforcements, they have dimensions that are approximately equal in all directions.

Nevertheless, particulate fillers have certain disadvantages due to its low strength compared to that of fibres. Besides, there is lack of directional property of the composite. Finally, they possess enhanced processing difficulty at high loading.

2.3 Natural Fibres

Natural fibres are one of the key renewable resource materials throughout the world, especially in the tropics. In various part of the world, there are about 2000 species of useful fibre plants used for many applications. In many developing countries, these locally grown fibres are used as alternative for expensive synthetic fibres. Natural fibres are divided based on their origins into vegetable, animal, and mineral. Vegetable fibres are composed of cellulose while animal fibres are made up of proteins (hair, silk and wool). Vegetable fibres can also be divided into smaller groups, based on their origin within the plant into bast, leaf and fruit/seed fibres. Sponge-gourd is classified as fruit fibre. In recent times, a number of investigators have been involved in looking into the utilization of natural fibres as load bearing constituents in composite materials. The application of such materials in composites has intensified due to their relative cheapness and their ability to recycle. This is in addition to specific strength that is higher than that of glass fibre reinforced composites and similar specific modulus (Bledzki and Gassan, 1999). Natural fibres can be considered as composites of hollow cellulose fibrils held together by a lignin and hemicelluloses matrix (John and Rajesh, 2008). It can be considered as a naturally occurring composite consisting mainly of cellulose fibrils embedded in lignin matrix. The cellulose fibrils are aligned along the length of the fibre, which render maximum tensile and flexural strengths, in addition to providing rigidity.

2.4 Sponge-gourd fibre

Luffa is derived from the cucumber and marrow family and originates from America. Luffa aegyptiaca commonly called Luffa cylindrica, sponge gourd, loofa, vegetable sponge, bath sponge or dish cloth gourd, is a member of cucurbitaceous family. Sponge-gourd is a lignocellulose material composed mainly of cellulose, hemicelluloses and lignin. The fruits of Sponge-gourd are smooth and cylindrical in shape. Mature Sponge will produce at least 30 seeds, while some will produce many more. It has alternate and palmate leaves comprising petiole. The leaf is 13 and 30cm in length and width respectively and has the acute end lobe. It is hairless and has serrated edges. The flower is yellow and boom in August- September. The fruit is green and has a large cylinder- like shape. The outside of the fruit has vertical lines and a reticulate develops inside of the flesh. It grows to about 12cm long. The stem is green and pentagonal and grows climbing other physical solids. The plant is cultivated in many countries, including Brazil, where its cultivation has an increasing economic importance (Srinivasan et al, 2014).

Sponge-gourd is an annual vine that quickly covers any support it finds. When it finds no support, it crawls along the ground. The leaves are large and lobed with silver spots on the top. The yellow stricking flower is about 5-7.5cm in diameter. They are cylindrical and smooth and slightly wider at the tip. The young fruits are small and look like cucumber. It is mind blowing to note that the number of species in the genus Sponge varies from 5 to 7. Only 2 species Luffa Cylindrica and ribbed or ridge gourd (Luffa acutangular) are domesticated. 2 wild species are Luffa graveolens and Luffa echinata. The main commercial production countries are China, Korea, India, Japan and Central America (Oboh and Aluyor, 2009).

Sponge-gourd, the fruit of luffa cylindrica (LC) is one of the most interesting candidates for reinforcing polymer matrix due to its high specific strength and modulus , low density of about 0.82 to 0.92 g/cm^3 which is lower than the density of some common natural fibres like Sisal of about 1.26 to 1.45 g/cm^3, Hemp about 1.48 g/cm^3, Coir about 1.25g/cm^3, Ramie about 1.5g/cm^3 and Cotton about 1.51 to 1.6 g/cm^3 (Siqueira et al, 2010), renewability and no health risk. In addition, Sponge-gourd is a tropical plant which can cultivate in a wide range of areas in Nigeria.

2.5 Structural subdivision of sponge-gourd

The structure of sponge-gourd is subdivided into four parts: outer surface, middle part, inner surface and core part. Fibre is the basic structural element of sponge-gourd. Therefore, to comprehend the mechanical behaviour of the entire sponge, an understanding of its structure is essential.

In an interesting study to advance an understanding of the structure – function relationship of sponge-gourd, Chen et al. (2014), investigated the sponge structure from a single fibre to a segment of the sponge cylinder. They reported that the stress-strain curves of the single fibres are nearly linear-elastic, and that the fibres failure strains are very small, around 5%. In terms of their basic materials constant, they reported that Young’s modulus and strength are about 2368MPa and 103MPa respectively. For single fractured fibre, they reported that it is around 0.4mm in diameter and that the orientation of cellulose fibrils is parallel to the longitudinal axis of the fibre. For block fibre samples, they reported that they have different mechanical behaviours, which depend on loading directions and locations in the sponge, where the samples were taken. They discovered that in general, the stress-strain curves demonstrated the typical three stages of the mechanical behaviour of porous materials which are: linear-elasticity (strain < 10%), plateau (10% < strain < 60%) and densification (strain > 60%). They concluded that in all, the existence of inner surfaces apparently enhances the structures' mechanical performance.

2.6 Chemical composition of Natural fibres

The reinforcing efficacy of natural fibres is associated with the nature of cellulose and its crystallinity, which in turn depends on the type of fibre. The main constituents of natural fibres are cellulose, hemicelluloses, and lignin. They are therefore referred to as lignocellulosic materials. In lower quantities, pectins, pigments and extractives can be found (Siqueira et al, 2010). The properties of each constituent contribute to the overall properties of the fibre.

2.6.1 Cellulose: The main constituent of any plant fibre is cellulose (Akil et al, 2011). It is the natural homopolymer (polysaccharides), where D-anhydroglucose ($C_6H_{11}O_5$) repeating units are connected to each other with β-1, 4-glycosidic linkages. Cellulose is a semi crystalline polysaccharide and is responsible for the hydrophilic nature of natural fibres. The degree of polymerization is about 10,000. Each repeating unit contains three hydroxyl groups. These hydroxyl groups and their ability to hydrogen bond play a key role in directing the crystalline packing and also govern the physical properties of cellulose. The crystal nature of naturally occurring cellulose is known as cellulose 1. Cellulose is resistant to strong alkalis (17.5 wt %) but is easily hydrolyzed by acids to water-soluble sugars. It is relatively resistant to oxidizing agents.

2.6.2 Hemicelluloses: Hemicelluloses found in the natural fibres is believed to be a compatibilizer between cellulose and lignin (Susheel et al, 2009). There exist differences between cellulose and hemicelluloses which include: Hemicelluloses contain several different sugar units whereas cellulose contains only 1,4-β-D-glucopyranose units. Besides, the latter exhibit a considerable degree of chain branching containing pendant side groups giving rise to its non crystalline nature, whereas the former is a linear polymer. Finally, the degree of polymerization of native cellulose is 10 to 100 times higher than that of hemicelluloses, which is about 50 to 300. The amorphous nature of hemicelluloses results in its being partially soluble in water and alkaline solutions. It is responsible for the biodegradation, moisture absorption, and thermal degradation of the fibre.

2.6.3 Lignin: This is a complex hydrocarbon polymer with both aliphatic and aromatic constituents. It is totally amorphous and hydrophobic in nature. It is insoluble in most solvents and cannot be broken down to monomeric units. Lignin is comprised mainly of aromatics and has little effect on water absorption Himadri and Dipul, (2014). Besides, it acts as protection against biological attack and a stiffener to give stem its resistance against gravity forces and wind. Lignin is thermally stable but is responsible for UV degradation.

Pectin, whose function is to hold the fibre together, is a polysaccharide like cellulose and hemicelllulose. Waxes consist of different types of alcohols.

The chemical composition of Sponge-gourd fibres depends on several factors, such as plant origin, weather conditions, soil nature etc. (Siqueira et al, 2010).

Chemical composition and physical properties of sponge-gourd fibres reported by some researchers are listed in Tables 2.2 and 2.3 respectively.

Table2.2 Chemical composition of sponge gourd fibres as reported by researchers

Holocellulose (%)	-- Cellulose (%)	Hemicellulose (%)	Lignin (%)	Extractives(%)	Ashes (%)	Reference
82.4	63.0	19.4	11.2	3.2	0.4	Tanobe et al, 2005
-	63.0±2.5	20.88±1.4	11.69±1.2	-	0.4±0.10	Saw et al, 2013
83 ± 1	65.5 ± 0.5	17.5 ±0.5	15.2 ±1.0	3.1 ± 0.5	0.7 ± 0.2	Siqueira et al, 2010

Table 2.3 Physical properties of sponge-gourd fibre as reported by researchers.

Density	Diameter	Aspect ratio	Mcrofibrillar Angle	Reference
0.92 ± 0.10 (gm/cm^3)	270 ± 20 (μm)	340 ± 5	12 ± 2	Saw et al, 2013
0.92 ± 0.05 (g/cm^3)	o.631 ± 0.217	-	-	Tanobe et al, 2005
0.82 to 0.92 (g/cm^3)	-	-	-	Siqueira et al, 2010

In an interesting study to determine the suitability of sponge-gourd in the preparation of cellulose whiskers and microfibrillated cellulose, Siqueira et al. (2010), performed sugar analysis of hydrolysis residue. From their result in Table 2.4, they concluded that glucose being the main sugar present in the residue confirms its cellulosic nature and indicates that sponge-gourd is a good source for the preparation of cellulose whiskers and MFC.

Table 2.4 Sugar Composition (wt %) of Sponge-gourd’s Hydrolysis Residue (Siqueira et al, 2010)

Arabinose	Galactose	Glucose	Xylose
0.74±0.01	0.58±0.07	67.22±2.57	12.26±0.14

2.7 Factors Affecting Fibre Reinforcement

When fibres are used as reinforcement they broaden the useful range of polymer properties. At the same time, they make the analysis of fibre reinforced rubber composite more complex. In his work, Dzyura (1980) discovered that there are many variables which affect the performance of a fibre reinforced rubber composite. They include:

2.7.1 Fibre Dispersion: It goes without saying that good fibre dispersion in the polymer matrix is one of the major requirements for obtaining a satisfactory performance from short-fibre composites. Good dispersion is an indication that the fibres are separated from each other. That is there are no clumps and agglomerates, and each fibre is surrounded by the matrix. In contrast, inadequate fibre dispersion will result in an inhomogeneous mixture of resin-rich areas which are weak and fibre-rich areas (i.e., clumps) which are prone to micro cracking. This is undesirable because micro cracks contribute to poorer mechanical properties of composites. To achieve maximum strength and performance of the composite material, it is imperative to ensure homogeneous fibre dispersion. It is worth mentioning that, according to Derringer (1971), two major factors that affect the degree of fibre dispersion are fibre-fibre interaction such as hydrogen bonding between the fibres and fibre length. Fibre length should not be too long, otherwise the fibres may get entangled with each other and not too short otherwise stress transfer area will be too small for the fibres to offer effective reinforcement. Some of the suggested methods to overcome poor fibre dispersion are fibre surface modification, use of dispersing agents, fibre pre-treatments and increased shear force and mixing time (Derringer, 1971).

2.7.2 Fibre-Matrix Adhesion: This factor plays a significant role in the reinforcement of composites with short fibres. When loads are applied, it is not applied directly to the fibres but to the matrix. The load must therefore be transferred well from the matrix to the fibres to have composites with outstanding mechanical properties. Strong and efficient fibre-matrix interface is required for this. Fibre matrix interface can be controlled by fibre surface treatment and by the use of additives.

2.7.3 Fibre aspect ratio: This has a considerable effect on composite properties hence it is important to preserve fibre length as far as possible during rubber composite processing. It is the length to diameter ratio of a fibre. It is important to note that for each short-fibre composite system, there is a critical fibre aspect ratio that may be defined as the minimum fibre aspect ratio in which the maximum allowable fibre stress can be achieved for a given load. For maximum reinforcement, the fibre aspect ratio of any composite should be above its critical value. This will guarantee maximum stress transfer to the fibres before the composite fails. If the fibre aspect ratio is lower than its critical value, inadequate stress will be transferred and reinforcement by the fibres will be inefficient, since the fibres are not loaded to their maximum stress value. Then again, if the fibre aspect ratio is too high, the fibres may get entangled during mixing and then hinder fibre dispersion. John and Rajesh (2008) concluded that fibre aspect ratio must be in the range of 100 – 200 for optimum effectiveness.

2.7.4 Fibre orientation: This is yet another important factor that influences the mechanical properties of short fibre composites. This is not unconnected with the fact that fibres in such composites are rarely oriented in a single direction, essential for the fibres to offer maximum reinforcement effects. Accordingly, the extent of reinforcement in a short-fibre composite is found to be strongly reliant on the orientation of each single fibre with respect to the loading axis. During the processing of short fibre composites, changes in fibre orientation take place continuously and progressively. These changes are related to geometrical properties of the fibres, the viscoelastic properties of the matrix, and the change in shape of the material which is produced by the processing operation.

2.7.5 Fibre volume fraction: The properties of a composite are strongly influenced by its composition. Variation of composite properties, particularly tensile strength, with fibre content can be calculated by using several models such as the ‘Rule of Mixtures’. The composition can be stated either using weight fraction (Wf), which is relevant to fabrication or using volume fraction (Vf), which is relevant in property computations. The volume fraction and weight fraction of the filler can be calculated with the equations below:

$$\mathrm{Wf} = \frac{weight\ of\ filler}{weight\ of\ composite} \qquad 2.1$$

$$\mathrm{Vf} = \frac{volume\ of\ filler}{volume\ of\ composite} \qquad 2.2$$

The corresponding fibre volume fraction in which the strength properties of the composite cease to decline with fibre addition , and begin to improve, is known as the optimum or critical fibre volume fraction, C_{crit} (Derringer, 1971). For short fibre composites to perform well during service, the matrix must be loaded with fibres beyond this critical value (Derringer ,1971).

2.8 Advantages and disadvantages of natural fibres

The emergent attraction in lignocellulosic fibres is chiefly due to their economical production with few necessity for equipment and low specific weight, which results in a higher specific strength and stiffness when compared to glass reinforced composites. Besides, they also present safer handling and working conditions compared to synthetic reinforcements. More so, natural fibres are nonabrasive to mixing and molding equipment, which can contribute to substantial cost reductions. Furthermore, natural fibre has positive impact on the environment. They are renewable resources, where they are biodegradable and their production requires little energy (Akil et al, 2011). Additionally, they are carbon dioxide neutral i.e. they do not return excess carbon dioxide into the atmosphere when they are burned. Moreover, their processing is environmental friendly, offering better working conditions and therefore, a reduction in risk of dermal or respiratory problems (ibid.). What's more, natural fibres possess high electrical resistance. Finally, an additional factor is their availability.

Nevertheless, despite the advantages mentioned above, the foremost disadvantage of natural fibre reinforced composites is the difficulty in compounding. This is attributed to the polar and hydrophilic nature of the fibres and the non-polar characteristics of the polymer matrix which causes poor wetting. Added to this is the processing temperature of composites which is limited to 200℃ because vegetable fibres degrade at higher temperatures. Thus, restriction is placed on the choice of matrix material. Additionally, natural fibres soak up high moisture, which leads to swelling and presence of voids at the interface. This results in poor mechanical properties and reduces dimensional stability of composites. More so, another constraint to the fruitful utilization of natural fibres for long-lasting composite application is low microbial resistance. They are easily attacked by a variety of organisms, at high humidity and temperature, leading to rot and mildew.

It is worthy to mention that the advantages of natural fibres overshadow the disadvantages. Besides, most of the shortcomings have corrective measures in the form of surface modification.

2.9 Surface modification of Natural fibres

The main problem of using natural fibre to reinforce NR is the incompatibility between a hydrophilic natural fibre and a hydrophobic NR, leading to poor mechanical properties of the composites. However, the interfacial adhesion between natural fibre and NR can be improved by different kinds of physical or chemical surface treatments such as corona discharge or reaction with alkyl ketone dimmers, alkalis, silane –coupling agents, etc. (Tanobe et al, 2004). The treatment of fibres is currently an area of research receiving significant attention. The absorption of water is commonly thought to occur at the free hydroxyl groups on the cellulose chains. With a ratio of 3 hydroxyl groups per glucose repeat unit, the amount of water that can be absorbed is substantial. By capping the hydroxyl groups, this ratio can be reduced.

2.9.1 Physical Methods

Physical methods include: stretching, calendering, thermo treatment and production of hybrid yarns (Himadri and Dipul, 2014). They do not change the chemical composition of the fibres. Physical treatments change structural and surface properties of the fibres and thereby influence the mechanical bonding to polymers. Another physical method is by Electric discharge which can be corona or cold plasma treatment.

2.9.2 Chemical Methods

Cellulose fibres exhibit a highly polar surface due to the presence of hydroxyl groups. These hydroxyl groups enable the formation of hydrogen bonds in the interphase of reinforced composite materials. But in order to get access to these hydroxyl groups, a cover of pectin and other waxy substances must be removed. On the other hand, the high polarity of the cellulose fibre surface is the reason for their hydrophilic behaviour, which induces fibre swelling. Intense surface treatment is therefore required for fibre cleaning and preparation for interfacial bonding. Chemical pretreatment of natural fibres is so effective in that it can clean the fibre surface, chemically modify the surface, stop the moisture absorption process and increase the surface roughness. Some important chemical treatments include treatment with alkali, silane, acetylation, isocyanates, etc. They are briefly discussed below.

2.9.2.1 Alkali treatment

Alkaline treatment, or mercerization, is one of the most used chemical treatments of natural fibres for reinforcement. The principal modification done by alkali treatment is the disruption of hydrogen bonding in the network structure, thereby increasing surface roughness. This treatment removes a certain amount of lignin, wax and oils covering the external surface of the fibre cell wall, depolymerizes cellulose and exposes the short length crystallites (Mohanty et al, 2001). Addition of aqueous sodium hydroxide (NaOH) to natural fibre promotes the ionization of the hydroxyl group to the alkoxide (Agrawal et al, 2000).

$$\text{Fibre} - \text{OH} + \text{NaOH} \rightarrow \text{Fibre} - \text{O}^{-}\text{Na}^{+} + \text{H}_2\text{O} \qquad 2.3$$

Consequently, alkaline processing directly influences the cellulosic fibril, the degree of polymerization and the extraction of lignin and hemicellulosic compounds (Himadri and Dipul ,2014). In other words, alkali solution influences not only the cellulosic components inside the plant fibre but also the non cellulosic components (hemicelluloses, lignin, and pectin) (John and Rajesh, 2008). In alkaline treatment, fibres are immersed in NaOH solution for a given period of time. The effect of alkali on cellulose fibre is a swelling reaction, during which the natural crystalline structure of the cellulose relaxes. Native cellulose shows a monoclinic crystalline lattice of cellulose-1, which can be changed into different polymorphous forms through chemical or thermal treatments. The type of alkali and its concentration will influence the degree of swelling, and hence the degree of lattice transformation into cellulose-11. Studies have shown that Na^+ has got a favorable diameter, able to widen the smallest pores in between the lattice planes and penetrate into them. Consequently, sodium hydroxide treatment results in a higher amount of swelling. Sodium hydroxide can cause a complete lattice transformation from cellulose-1 to cellulose-11(which is thermodynamically more stable than cellulose-1) in contrast to other alkalis that produce only partial lattice transformation (John and Rajesh, 2008).

2.9.2.2 Silane Treatment

Silane is a chemical compound with the chemical formula SiH_4. They are hydrophilic compounds with different groups attached to silicon such that one end will interact with matrix and the other end can react with hydrophilic fibre, which act as a bridge between them. The uptake of silane is very much dependent on a number of factors including hydrolysis time, temperature, pH etc. hydrocarbon chains provided by the silane restrain the swelling of the fibre by creating a cross-linked network, due to covalent bonding between the matrix and the fibre. The reaction schemes are given as follows (Himadri and Dipul, 2014).

$$CH_2CHSi\,(OC_2H_5)_3 \xrightarrow{H_2O} CH_2CHSi(OH)_3 + 3C_2H_5OH \qquad 2.4$$

$$CH_2CHSi(OH)_3 + \text{Fibre} - OH \rightarrow CH_2CHSi(OH)_2O - \text{Fibre} + H_2O \qquad 2.5$$

2.9.2.3 Acetylation of natural fibres

Acetylation describes a reaction that introduces an acetyl functional group (CH_3COO^-) into an organic compound. It is a well known esterification method causing plasticization of cellulosic fibres. The treatment stabilizes the cell walls against moisture, improving dimensional stability and environmental degradation. Pretreatment of fibres with acetic anhydride substitutes the polymer hydroxyl groups of the cell wall with acetyl groups, modifying the properties of these polymers so that they become hydrophobic (Hill et al, 1998). The reaction involves the generation of acetic acid (CH_3COOH) as a byproduct which must be removed from the lignocellulosic material before the fibre is used (Himadri and Dipul, 2014). Hydroxyl groups that react with the reagents are those of lignin and hemicelluloses (amorphous material), whereas the hydroxyl groups of cellulose (crystalline material) are being closely packed with hydrogen bonds, prevent the diffusion of reagent and thus results in very low extents of reaction (Susheel et al, 2009). Acetic anhydride reacts with more reactive hydroxyl groups as per the reaction shown below.

$C_6H_7O_2(OH)_3 + 3(CH_3CO)_2 \rightarrow C_6H_7O_2(CH_3COO)_3 + 3(CH_3COOH) + 54Kcal$ 2.6

2.9.2.4 Isocyanate Treatment

An isocyanate is a compound containing the isocyanate functional group

-N=C=O, which is highly disposed to reaction with the hydroxyl groups of cellulose and lignin in fibres and forms strong covalent bonds, thereby creating better compatibility with the binder resin in the composites.

It is important to note that sustainability and end of life after use are important considerations to make when developing polymer composites from renewable resources. These considerations include the toxicity and environmental impact of using various chemical or physical methods for improving the properties of these materials. It is interesting to note that some chemical techniques may be toxic. This necessitated the warning by Himadri and Dipul (2014), that since isocyanates are carcinogenic, their use may not be feasible for the development of polymer composites from renewable resources.

2.10 Carbonization of Natural fibres

Activated carbon was first introduced industrially in the first part of the 20th century, when activated carbon from vegetable mineral was produced for use in sugar refining. Powdered activated carbon was first produced commercially in Europe in the 19th century, using wood as a raw material, which found wide use in the sugar industry. In the US, the first production of activated carbon used black ash as the source, after it was accidentally discovered that the ash was very effective in decolorizing liquids (Karthikeyan et al, 2007).

According to (Hall et al, 1982), biomass can be converted to energy and fuels through thermochemical and biochemical processes. It is interesting to note that carbonization is one of the thermochemical processes that convert biomass into charcoal, a domestic fuel for cooking in the rural areas. Hugh (1990), defined carbonization process as the step in which the organic material is heated slowly in an inert environment at temperature of 450℃ - 500℃, which may extend to 1300℃. During the process, the organic material is changed into a material that is fundamentally carbon. On his part, Hassler (1963) concluded that carbonization is the production of charred carbon from a source material. According to him, the process is generally accomplished by heating the source material usually in the absence or limited amount of air to a temperature sufficiently high to dry and volatilize substances in the carbonaceous material. In his work, Walter (1985) concluded that carbonization process is complex and that several reactions may take place concurrently such as dehydration, devolatilization, and condensation.

It goes without saying that the physical properties of carbon are significant in determining which form is best suited for specific application. For instance, abrasion resistance or hardness of activated carbon will be important if the carbon is to be used in application where recurrent back washing is required. Iodine value is an important characteristic of activated carbon, as it gives a measure of the micro pore volume of carbon and it approximates the total internal surface of the carbon. The ash level reflects the purity of the carbon (Yusufu et al, 2012).

2.11 Properties of natural fibre / natural rubber composites

The performance of materials, are presented in terms of their mechanical characteristics such as tensile, flexural, compression, impact and wear properties. These characteristics are essential to determine material ability, especially under extreme and critical conditions, which are directly connected with engineering performance. For the past few years, some studies have been performed on sponged-gourd fibre and other natural fibre reinforced composite, in order to fully characterize their mechanical behaviour.

2.11.1 Effect of sponge-gourd fibre preparation method

The mechanical properties of composites are dependent on the properties of the fibre and preparation of composites. In an interesting study, Paglicawan et al. (2005), investigated the mechanical properties of composite as a function of different methods of sponge-gourd fibre preparation. Different methods of preparation of the fibre were used in the fabrication of the polymer-bonded composite. The first sample was cross-sectioned of sponge-gourd fibre in its original shape. The second sample was cut lengthwise with middle fibrous skeletal and in the third sample, the middle portion of the fibrous skeletal was removed, forming chopped fibres. The mechanical properties of the composite in tension was determined along longitudinal and transverse directions for samples 1and 2, while sample 3 was presumed to be isotropic material and only one direction was taken. The result showed that all the samples regardless of preparation and number of piles in the composites resulted to higher tensile properties in transverse direction. For sample three where sponge-gourd was chopped into small segments, the composites produced have low tensile strength but high modulus. With the addition of sponge-gourd fibre, the tensile strength of the composites increased but the tensile modulus decreased.

They also determined the mechanical properties of the composite in flexure, and reported that generally, the flexural strength of the composite were almost the same despite varying the fibre preparation except in transverse direction of sample 1. The stiffness of composite varied depending on the sample preparation and direction of loading.

2.11.2 Effect of chemical modification

Chemical modification is one of the methods used in improving the compatibility between hydrophilic natural fibre and hydrophobic polymer matrix. Siqueira et al. (2010) carried out a study on luffa cylindrica as a lignocellulosic source of fibre, microfibrillated cellulose (MFC), and cellulose nanocrystals. The fibres were treated with 4wt% NaOH solution at 80℃ for 2h under mechanical stirring. For whiskers, acid hydrolysis was achieved at 50℃ with 65wt% sulphuric acid (pre-heated) for 40 min, using mechanical stirring. They reported that NaOH and bleaching treatments lead to fibre fibrillation and breaking-down of the fibre bundles into smaller bundles. Besides, X-ray diffraction was used to calculate the crystallinity index (I_c) of the various samples. They reported an increase in the degree of crystallinity in the treated samples, which they attributed to the partial elimination of amorphous regions from the fibres during alkali and bleaching treatments, performed before MFC and whiskers preparation.

In yet another interesting study, the effect of chemical treatment on the properties of sponge-gourd fibre reinforced epoxy composites was investigated by Saw et al.

(2013). In their work, two chemical treatments, alkalization (2 h agitation with 5% NaOH) and furfurylation (graft furfuryl alcohol followed by oxidation with (1N) $NaClO_2$ solution), were conducted on sponge-gourd fibre surfaces. They reported that NaOH treatment disrupts hydrogen bonding in the network structure, thereby increasing surface roughness. According to them, this treatment removes a certain amount of lignin, wax, and hemicelluloses of the fibre cell wall, depolymerizes cellulose, and exposes the short-length cellulose fibril. The ionization of hydroxyl groups of fibres into alkoxide increases the fibre strength and stiffness and hence, increases mechanical properties. They determined the weight changes of the fibre gravimetrically after alkali treatment and observed a weight loss near 2%. On the effect of chemical treatment on the surface composition of sponge-gourd fibre, they observed significant differences in the carbon (C) and oxygen (O) percentages, between untreated and treated fibres. The O/C ratio for untreated fibre was 0.58, which is an indication that there was still lignin on the surface. When this ratio is 0.83, the sample is pure cellulose, whereas pure lignin if in the range of 0.31 to 0.40. They concluded that the value was increased to 0.66 for alkali-treated fibre. This they attributed to the removal of some parts of lignin and hemicellulose content and an increase in cellulose content. The ratio was 0.67 for oxidized sponge-gourd fibre and a maximum of 0.71 for FA-grafted fibre. Tanobe et al. (2004) made the same observation on Brazilian sponge-gourd fibre with an O/C ratio of 0.57. In terms of mechanical properties, they reported that the tensile properties of the composite were influenced by the strength and modulus of the fibres. The extent of improvement in mechanical properties for FA-grafted fibre reinforced composites was higher than that observed for alkali treated fibre composites. On impact energy, the chemically treated fibre composites had higher values of impact strength than untreated fibre composites. More so, it was also reported that the rate of water absorption was lower for treated composites compared to untreated sponge-gourd based composites after 240h exposure in water. It was concluded that untreated fibre based composites exhibit higher rates of water absorption due to the hydrophilic nature of lignocellulose, as well as due to the capillary action in the matrix when fibre composites are exposed to water. This could be possible due to a higher porosity or to the existence of voids formed during processing that could accelerate the diffusion. Reduction in water uptake was attributed to good interfacial contact between fibre and matrix, and because the fibres had become more hydrophobic.

On their part, Tanobe et al. (2004), attempted an interesting study on comprehensive characterization of chemically treated Brazilian sponge-gourds. In their work, different chemical treatments were conducted on the fibres with aqueous solutions of NaOH 2%, or methacrylamide (1-3%) at distinct treatment times. They reported that treatment with NaOH was found to adequately modify the fibre surface in preparation for its use as reinforcement of composite materials, not causing severe damage to the fibre unlike treatment with methacrylamide which severely damaged the fibre. In terms of effect of treatment on thermal degradation, they reported that treated fibres showed a slight increase in thermal resistance. Temperatures higher than 250℃ showed the largest difference although, the fibre at this point has undergone severe thermal degradation.

The effect of fibre surface treatments on the tensile and water sorption properties of polypropylene – sponge-gourd fibre composites was studied by Demir et al. (2006). To improve the interaction between matrix and fibres, silane coupling agents, namely, aminopropyltriethoxy silane and mercapto silane were employed in the pretreatment of sponge-gourd fibre. Maleated PP was used for the improvement of surface of the PP matrix. The tensile properties of the untreated and treated composites were determined as a function of filler loading. In terms of filler loading, they reported that, in general, the tensile strength of the treated and untreated composites decreased as the fibre content increased. This was as a result of the reduction in the effective matrix cross section. The observed reduction in tensile strength of PP composites was also attributed silane-coupling agents. However, the decrease was greater in the untreated composites than the treated composites. Besides, they reported an increase in tensile strength on employment of coupling agents. This was attributed to better adhesion between the filler and the matrix. More so, the Young’s modulus of the composites as a function of filler content for the different treatment conditions, increased as the fibre loading increased. This was also attributed to better adhesion between the fibre and the matrix by chemical interactions. Better adhesion yields to more restriction to deformation capacity of the matrix in the elastic zone increasing Young’s modulus. In terms of elongation at break, it was reported that elongation at break values for all composites decreased with the increase in fibre loading. Furthermore, with the aid of SEM, they studied the effect of surface treatment on the interface between the PP and sponge-gourd fibres, by examining the fracture surfaces of the tensile tested composites. They reported that there were voids between fibre and matrix surface for untreated composites which is an evidence of poor adhesion. This seems to facilitate debonding of the fibre. SEM micrographs of treated composites indicated that treatment facilitates good adhesion between fibre and matrix. On the contrary, a decrease in water absorption with treatment was reported. Water absorption in cellulose fibres is caused by hydrogen bonding between free hydroxyl groups on cellulose molecules and water molecules. The coupling agents form hydrogen or covalent bonds with some of free hydroxyl groups of cellulose, which reduce the water absorption capacity of cellulose. Another reason for the decrease was better adhesion between the fibre and the matrix, which resulted in a decrease in voids between fibre and polymer matrix. Poor adhesion causes cracks and voids between the matrix and fibre which causes easy penetration and storage of water through the voids.

On their part, Boynard et al. (2003), studied the flexural properties of sponge gourd fibre-reinforced polyester composites. Composites were prepared from sponge gourd fibres treated with NaOH of different concentrations, viz. 5, 10, 20, 40, and 60% at room temperature. Composites were also prepared with fibres treated with 1 and 5% NaOH at 100℃. The authors reported that mercerization treatment produced strong morphological changes on the surface of sponge gourd fibres. According to them, the outer surface of the fibres was completely removed exposing the inner fibrilla surface. Composites prepared from sponge fibres treated with 5% NaOH exhibited the best flexural properties. Flexural modulus increased by 14% when compared with composites containing untreated fibres. In their conclusion, they attributed the increase to the mechanical interlocking due to increase in fibre roughness and contact area of fibres.

2.11.3 Effect of filler loading

The properties of a composite are strongly influenced by its composition.

In an interesting study, Darinya et al. (2012) investigated the effects of defatted rice bran as a filler for natural rubber vulcanisate. On cure time, they reported that the addition of fillers decreased the cure time of the rubber compounds. In terms of tensile strength, they reported a decrease in its value, because of the inability of the fillers to support stress transferred from the rubber matrix. At a similar filler loading, clay gave the highest tensile strength, which corresponds to its highest surface area, since surface area is the most important factor controlling the tensile strength. On tear strength, they observed that the addition of fillers somewhat reduced the tear strength of the vulcanisate. They concluded that the results was an indication that the tear strength seemed to be affected by the surface area of the fillers and also probably by a low rubber-filler interaction. In terms of hardness, they reported that hardness increased on addition of filler. They concluded that improvement in hardness was caused by an increase in the cross-link density of the vulcanisates. On the contrary, they reported a slight decrease in the resilience of the vulcanisates i.e. the elasticity of the rubber chains with addition of filler. They also investigated the abrasion resistance of the composite. They concluded that the defatted rice bran filled vulcanisate exhibited highest abrasion resistance. In their opinion, it corresponded to its highest hardness and cross-link density.

In yet another interesting study, Fidelis et al. (2013) investigated the curing and physico-mechanical properties of maize stalk filled NR composite as a function of filler loading. They reported that in terms of cure characteristic, the torque value (an indication of crosslink density and degree of reinforcement of filler in the matrix) increases with increase in filler loading. An indication that as more and more filler gets reinforced into the matrix, the mobility of macromolecular chains of NR becomes reduced resulting in more rigid vulcanisates. Thus, increase in torque with increasing filler loading is attributed to the increase of the crosslink density. In terms of scotch time, they reported a decrease in scotch time with increasing filler loading. The decrease in scotch time was due to the restriction of mobility and deformation of the matrix, with the introduction of mechanical restraints. However, the formulation with modified maize stalk and commercial filler had higher scotch time than the unmodified maize stalk due to good dispersibility of the modified and commercial filler in the rubber matrix. On cure time, they reported that the cure time of NR composites decreased with increasing filler loading. This was attributed to the higher time the rubber compounds remained in the mill during mixing. As the filler loading increases, the time of incorporation also increases and consequently generates more heat due to friction. They also reported that the introduction of unmodified maize fibre produces a rubber composite with lower Tensile strength compared to the NR. Again, at loadings below 10phr, the maize treated rubber composite has a lower Tensile strength than the NR. But this increased at loadings from 10 to 25phr, and then decreased at above 25phr. The decrease in Tensile strength above 25phr is due to high fibre population leading to fibre agglomeration resulting in difficult stress transmission from matrix to fibre and disturbance of the continuity of the matrix phase. They were of the opinion that the decrease in Tensile strength indicate the loading at which the filler bears more of the stress and it somehow shows the limiting loading levels. On Elongation at break, they reported that the values of Elongation at break (EAB) decreased with the increase in loading for all the fillers. They were of the view that a decrease in elongation at break was due to adherence of the filler to the rubber polymer matrix leading to the stiffening of the polymer chain and hence resistance to stretch when the strain is applied.

In another study, Ismail et al. (2002), investigated the effects of filler loading and bonding agents on the properties of Bamboo fibre filled natural rubber composites. They reported that the Mooney viscosity of the composites with and without bonding agent increased with increasing filler loading and the addition of bonding agent. This indicated that the addition of both filler and bonding agent increased the stiffness of the composites. They also reported that the effect of filler loading on Tensile strength and Tear strength of the composites was that of a decrease with increasing filler loading. This is however contrary to the result obtained by researchers like Derringer (1971), O’Connor (1977) and Setua and De (1983) who reported increased tensile strength with increased fibre loading. The effect of filler loading on tensile modulus and Hardness of the composites was also reported. They discovered that both properties increased with increasing filler loading.

However, with the presence of bonding agent, these properties were further increased as a result of better interaction between fibre and rubber matrix.

More so, an attempt was made to study the effect of filler loading on the mechanical properties of NR filled with Guinea Corn Husk by Tenebe et al. (2013). They reported an increase in tensile strength and modulus. This they attributed to high surface area of guinea corn husk suggesting better polymer filler interaction and hence enhanced better tensile properties. They also reported that elongation at break (EAB) decreased with increasing cross-links of the mixes for all fillers below 20phr, above which there was a gradual rise in the value of EAB. This was attributed to the spacing between cross-links which put more of the stress on a relatively few of the network chains. The highly stressed chains break first and their loads are distributed to other chains forcing them to either break or slip so as to relieve the stress on them. Another reason is adherence of the filler to the polymer phase leading to the stiffening of the polymer chain and hence resistance to stretch when the strain is applied. On hardness, they reported an increase with increasing filler loading. A decrease in the values of flex fatigue with increasing filler loading was recorded. This was attributed to 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. Besides, a decrease in the values of compression set with increasing filler load was credited to the degree of filler dispersion and its particle size which enhanced its performance. Finally, an increase in the values of abrasion resistance with increasing filler loading was credited to the degree of dispersion of the fillers.

2.11.4 Effect of carbonization

The mechanical properties of NR vulcanizates are greatly influenced by filler carbonization temperature and loading (Ekebafe et al, 2010).

In an interesting study, Aguele et al. (2014) carried out a comparative study of physical properties of polymer composites reinforced with uncarbonised (UC) and carbonized (CC) coir. Carbonization of coir was done at a temperature of 800℃ . It was later used as reinforcing filler in NR to obtain the vulcanizate. The authors reported that the UC coir induced higher reinforcement properties on the rubber composites than the CC. This was attributed to factors like particle size, structure and nature of filler. They observed that the UC and CC exhibited different levels of reinforcement potentials. They reported a general trend of increased hardness, tensile strength and modulus as filler quantities in the composites were increased. But, carbon black filled composites had higher values for each case. For compression set, elongation at break and flex fatigue, they reported a decrease in value as filler volume was increased. But carbon black filled samples had the least values for each case of filler volume. On the effect of carbonization on the filler properties of coir, the authors reported that carbonization decreased the reinforcement potential of coir as evidence in the performance of carbonized coir when compared with that of uncarbonized coir. They however attributed the decrease to the excessive carbonization temperature used during the study.

In another interesting study, Ayo et al. (2011) investigated the effects of carbonization temperature on the filler properties of groundnut shell powder. The authors reported that bulk density and percentage loss on ignition decreased with increase in carbonization temperature. They also revealed a gradual increase in iodine adsorption number as the carbonization temperature increased, whereas the filler pH changed from acidity to alkaline with increasing carbonization temperature.

In another study, the cure characteristics and physico-mechanical properties of carbonized bamboo fibre filled NR vulcanizates was investigated by Onyeagoro (2012). Bamboo fibres were carbonized at 600℃ for 3h and used as particulate filler in NR vulcanizates. On the effect of carbonization temperature on the filler, the author reported an increase in loss on ignition and iodine adsorption number and a decrease in pH. On the effect of filler loading and particle size on mechanical properties of vulcanizates, Ismail et al. (2002) reported a decrease in the values of tensile strength with increasing filler loading. This decrease was attributed to filler agglomeration due to poor dispersion. Following increase and decrease in tensile strength as reported by different authors cited in this review work, it is therefore a truism to say that for increase in the value of tensile strength to be obtained, one must ensure proper dispersion of filler in the matrix. This is however a function of how fine the filler is. More so, the author reported an increase in the tensile modulus and hardness with increase in filler loading.

The characteristics of activated carbon depend on the physical and chemical properties of the raw materials as well as activation methods used (Guo and Lua 2001). In an interesting study, Yusufu et al. (2012) investigated the influence of mode of activation on quality indices of activated carbons from selected local raw materials (cattle bone, coconut-shell and wood carbon). They also determined the optimum condition for carbonization and activations. The authors reported that increase in carbonization time resulted in increased weight loss for all the carbon samples and then a reduction after the maximum values were obtained. According to Oyoh and Igbokwe (2001), the reduction in weight loss after the maximum values is an indication of the beginning of activation of the already formed carbon. On iodine number, the authors reported that the number increased at higher activation temperatures, an indication that activation temperature had a significant influence on the iodine number. Ajayi and Olawale (2009), had the same experience when they attempted to produce activated carbon from Canariun

Schweinfurthi Nutshell using chemical process. The authors further observed that at activation temperature around 950℃ and above, the iodine number decreased. The authors were of the opinion that the optimum activation temperature is around 850 - 900℃ for phosphoric acid activation. Following their result, they concluded that acid was the best activating agent for both bone and coconut-shell carbon while heat is the best activating agent for wood carbon. It is interesting to note that, according to the authors, heat activated carbons show higher hardness than the acid activated carbons. This indicates that higher temperature treatments have significant effect on the hardness of carbon materials. This high hardness indicates high abrasion resistance which could be taken advantage of in applications where frequent back washing is required.

Both mechanical properties and thermal properties of composites are governed by molecular interactions between fibre and matrix. The more the interaction between fibre and matrix, the more will be the mechanical strength of the materials as well as more will be resistance to heat flow (Smita et al, 2004). The thermal degradation of natural fibres is a two-stage process, one in the temperature range 220-280℃ and another in the range 280-300℃. The low temperature degradation process is associated with degradation of hemicelluloses whereas the high temperature process is due to lignin (Saheb and Jog, 1999). An analysis of thermal properties of luffa cylindrica - resorcinol composites was attempted by Parida et al. (2014).

The authors studied the effect of incorporation of LC fibre (untreated and treated) on the thermal stability of the composites by observing the thermal degradation of composites. They reported that when alkali treated LC fibres were used in the composites, there were considerable changes observed in the thermal behaviour of the composites. For instance, DTG curve for treated fibre composite was shifted towards higher temperature region. Cellulose degradation temperature was reported to have shifted from 341.92℃ for composites with untreated fibre to 400℃ for samples with treated fibre. This they attributed to the exposure of higher amount of cellulose on the surface of the fibre caused by chemical treatment with alkali. This lead to better and enhanced interlocking between treated LC fibre and matrix. They also reported that the lignin degradation temperature was decreased from 579℃ for untreated fibre composites to 572℃ for treated fibre composites. This was attributed to partial removal of lignin due to chemical treatment of LC fibre. Due to increase in hydrophobicity of treated LC fibre, the % of moisture was reduced in the treated fibre composites. Decrease in thermal stability of untreated LC fibre composite is due to poor adhesion between the untreated LC fibre and the matrix. But when treated fibres are reinforced in the matrix, the thermal stability is enhanced indicating better adhesion between treated LC fibre and matrix.

In another interesting study, Tanobe et al. (2005) observed the thermal behaviour of untreated and treated sponge-gourd fibre. Following TGA analysis of the sample, the authors reported that the fibre mass decreased from about 93 (at 100℃) to 89 (at 250 ℃) and to 32% (at 350 ℃). The different regions were associated with the loss of retained water at 100℃, hemicelluloses degradation in the 200-260℃ region, cellulose degradation at 240-350 ℃ and lignin degradation at 280-500℃. The authors observed that between 100 and 250℃, degradation turned the lignocellulosic fibre into a brownish colour material, losing its strength, while at higher temperatures, up to 500 ℃, carbonization occurred with accentuated loss of material. The degradation reactions of lignin and cellulose became exothermic at about 270 and 300 ℃ respectively. According to them, pyrolysis of α-cellulose occurred at about 300 ℃ and that of lignin at about 400℃, while hemicellulose decomposed at lower temperature. The authors concluded that treated fibres showed slight increase in thermal resistance and that temperatures higher than 250 ℃ showed the largest difference, although at this point, it has undergone severe thermal degradation.

CHAPTER THREE

MATERIALS AND METHODS

3.1 Materials

The polymer material used in this study was natural rubber (NR). Natural rubber crumbs conforming to NSR – 10 (Nigeria Standard Rubber) was obtained from Rubber Research Institute of Nigeria (RRIN), Iyanomo, Benin City. This rubber grade also conforms to TSR – 10 (Technically Specified Rubber).

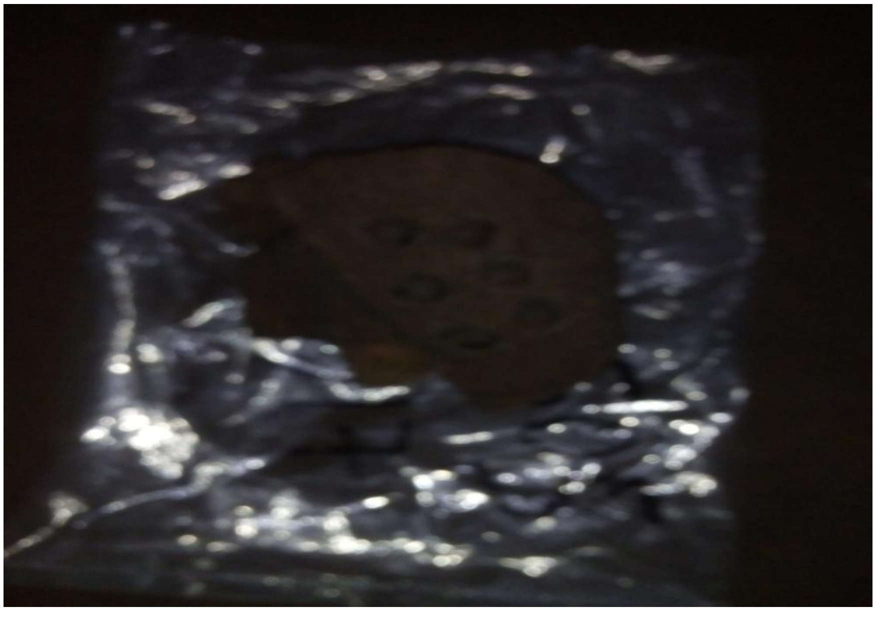

Figure 3.1: NSR-10

Sponge – gourd (Luffa Cylindrica) was obtained from Nkwere in Imo State and Ekosodin in Edo State. Carbon black (HAF N330) was obtained from Warri refining and Petrochemical Company. The carbon black was used as control in this study. Other compounding ingredients and additives used are of chemical grade and commercially available and were obtained from various chemical out lets in Benin City, Edo state. All reagents were used as obtained without further purification.

Figure 3.2: Fresh sponge-gourd

Figure 3.3: Dried sponge-gourd

Figure 3.4: Carbon black

3.2 Methods

3.2.1 Preparation of the fillers

The Pericarp (skin) of the ripe and dried fruit was removed. The Sponge – gourd was cut longitudinally and the seeds removed. It was then washed with deionized water to remove the adhering dirt, and thereafter divided into two parts. One part to be used as the uncarbonized sponge – gourd (US) was dried in an oven at 125℃ for 12 hours to remove moisture. It was then milled to fine powder using electrically powered plate grinding machine and sieved through a mesh size of 150 μm. The fine particles that passed through the mesh were collected as uncarbonized sponge – gourd powder. The other part to be used as carbonized sponge – gourd (CS) was sundried, weighed and then carbonized at a temperature of 500℃ (Tanobe et al, 2005). The carbonized sponge were then milled to a fine powder and sieved through a 150μm sieve mesh. The fine powder that passed through the mesh was then collected as carbonized sponge – gourd powder.

3.2.2 Characterization of sponge – gourd Fillers

In this study, the parameters investigated are: moisture content, loss on ignition, density, pH, particle size, and iodine adsorption number. These parameters are the determining factor in the level of distribution and dispersion of fillers in the rubber.

3.2.2.1 Moisture Content

To determine the moisture content of fillers, Moisture analyzer (AND ML – 50 Model) was used. During the process, 2g of carbonized sample was weighed and placed on the analyzer, at a temperature of 105℃ for 4minutes. The process was repeated for the uncarbonized sample, weighing 2g of it at 105℃ for 4minutes.

Figure 3.5: Moisture analyzer

3.2.2.2 Loss on Ignition

Loss on ignition refers to the mass loss of a combustion residue whenever it is heated in an air or oxygen atmosphere to high temperature. Determination of loss on ignition of the samples was done as described by Aguele et al. (2014). A sample of the filler was placed in a crucible and weighed. Then the crucible and content were placed in a furnace maintained at 500℃.

The loss on ignition was obtained as expressed in equation 3.1

$$\text{Loss on Ignition \%} = \frac{Weight\ loss}{Weight\ of\ sample} \times 100\ \% \qquad (3.1)$$

3.2.2.3 Filler Density

Filler density was determined by the procedure described by Gafar et al. (2012). In this method, 10g of carbonized and uncarbonized sample were measured in a pre-weighed measuring cylinder respectively. The weight of the cylinder and its content were measured. The weight of the sample was then obtained by subtracting the weight of the cylinder from the weight of the sample and cylinder. The specific density of filler was obtained using equation below:

$$\text{Density of filler} = \frac{Y_1 - Y_o}{V_o} \qquad (3.2)$$

Where Y_1 = weight of empty measuring cylinder + sample, Y_0 = weight of measuring cylinder, V_0 = volume of sample used.

3.2.2.4 Determination of Filler pH

The pH of the filler was determined according to ASTM D1512. pH meter 3510 Jenway Model was used for this purpose. A predetermined quantity (5g) of the filler was measured into a beaker. Then 25ml of deionized water was added. This was stirred vigorously and allowed to stand for 30minutes. The electrode of the pH meter was then inserted into the slurry so formed to obtain reading directly.

Figure 3.6: pH meter

3.2.2.5 Particle Size

The sponge – gourd powder was passed through a sieve of 150μm mesh. The particles that passed through the mesh were taken as having particle size of 150μm.

3.2.2.6 Determination of Iodine Value

The method used for surface area measurement of the fillers is iodine adsorption number (AWWA B 600-78, 1978). 2g of carbonized and uncarbonized samples were weighed respectively. They were then transferred to a dry, glass stoppered 250ml Erlenmeyer flask. 10ml of 5% HCl was added to the flask and swirled until wetting of the sample occurred. The flask was placed on a hot plate, contents brought to boil, and allowed to boil for exactly 30 seconds. The flask and contents were allowed to cool to room temperature. Then with pipette, 100ml of 0.10N iodine solution was added. The flask was vigorously shaken for 30 seconds. This was then immediately filtered through a filter paper. The filtrate was stirred in a beaker with a glass rod. Thereafter, with a pipette, 50ml was transferred into a 250ml flask. The 50ml sample was titrated with 0.10N sodium thiosulphate solution until the yellow colour almost disappeared. 1ml of starch solution was added and titration continued until the blue indicator colour disappeared. The volume of sodium thiosulphate solution used was recorded. The iodine number I_n was calculated using the equation below:

$$I_n = \frac{X\,A}{M} \qquad (3.3)$$

where

X = mg of iodine adsorbed by the sample

i.e. $X\ (12.693N_1)\ -\ (279.246N_2V)$

N_1 = normality of iodine solution

N_2 = normality of sodium thiosulphate solution

V = volume of sodium thiosulphate solution in ml

M = mass of sample in g

A = correction factor, depending on the residual normality N_T of the filtrate

$$N_T = N_2\,\frac{V}{50}$$

3.2.3 Compounding of Natural rubber composite

The filler loadings used in this work were varied from 3 to 20 phr (parts per hundred parts of rubber), for both carbonized and uncarbonized sponge. A separate set of composites was also prepared using carbon black as filler and also varied within the same range of filler loading. The rubber was masticated and mixed with the additives using a laboratory size two-roll mill according to the mixing cycle shown in Table 3.2. Mixing was done according to ASTM-D3184-80 (1983). The temperature of the mill was maintained at 70℃, to prevent cross-linking during mixing. Curing was done at a temperature of 130℃ for 5minutes.

The formulation used in preparing the natural rubber composites are shown in the Table 3.1 below

Table 3.1: Formulations for Reinforced Natural Rubber Composites.

Ingredient	**Content (phr)**
Natural rubber	100
Zinc oxide	5.0
Stearic acid	2.0
Trimethylquinoline (TMQ)	1.0
Mercarpto benzothiazole (MBT)	1.0
N-Cyclohexylbenzothiazole sulphonamide (CBS)	2.0
Sulphur	2.0
Processing oil	2.0
Fillers (US, CS and CB)	3 – 20

Table 3.2: Mixing steps and Mixing time

Mixing steps	Time (Minutes)
Natural rubber mastication	5
Addition of Stearic acid	1
Addition of Zinc Oxide	1
Addition of filler	10
Addition of MBT	1
Addition of TMQ	1
Addition of CBS	1
Addition of Processing Oil	1
Addition of Sulphur	2
Total	23

3.2.4 Measurement of Vulcanizate Properties

3.2.4.1 Determination of Tensile properties

Tensile properties were determined on a Zwick/Roell tensile tester model 2008 with a cross head speed of 200mm/min using dumb bell test pieces measuring 30mm×4mm×3.2mm according to DIN 53504,05/1994 test standard. The tensile strength at break was calculated as the elongation at break.

3.2.4.2 Determination of Hardness strength

The shore A hardness was obtained with a Durometer in accordance with ASTM-D 676-52T. The procedure involved placing the equipment on the dumb bell flat test pieces and recording the hardness when a little load was applied. This procedure was repeated for each sample and the average value taken.

3.2.4.3 Determination of Compression set

Compression set test was done in accordance with the ASTM-D 385. The test include a constant load (Method A) and constant deflection (Method B) as a compressive means. Compression set was determined by method A for various samples using a disc sample by placing the samples in the compression parallel plate and the samples were compressed to the spacer thickness retained after 24 hrs with force of 1.8KN at a temperature of 45℃. The compression set (CS) in the samples was calculated by the difference in the original thickness of the specimen and the thickness after distortion has been removed and allowed to recover for 30 minutes.

The compression set was calculated using the equation below.

$$\text{Compression Set \%} = \frac{T_{O-}T_f}{To} \times 100 \qquad (3.4)$$

where

T_0 = Original thickness

T_f = Final thickness

3.2.4.4 Determination of Abrasion resistance

Abrasion resistance is the ability of a material to resist mechanical action such as rubbing, scraping, or erosion that tends progressively to remove material from its surface. In this study, Wallace Akron tester was used in accordance with BS903. The procedure involved positioning the rubber test piece against an abrasive cylinder under constant speeds and held against the abrasive wheel by a constant force. The abrasion resistance was calculated from Equation 3.5

$$\text{Abrasion Resistance Index (ARI)} = S/T \times 100 \qquad (3.5)$$

where S = Standard Sample Volume Loss

T = Volume Loss for Test Sample

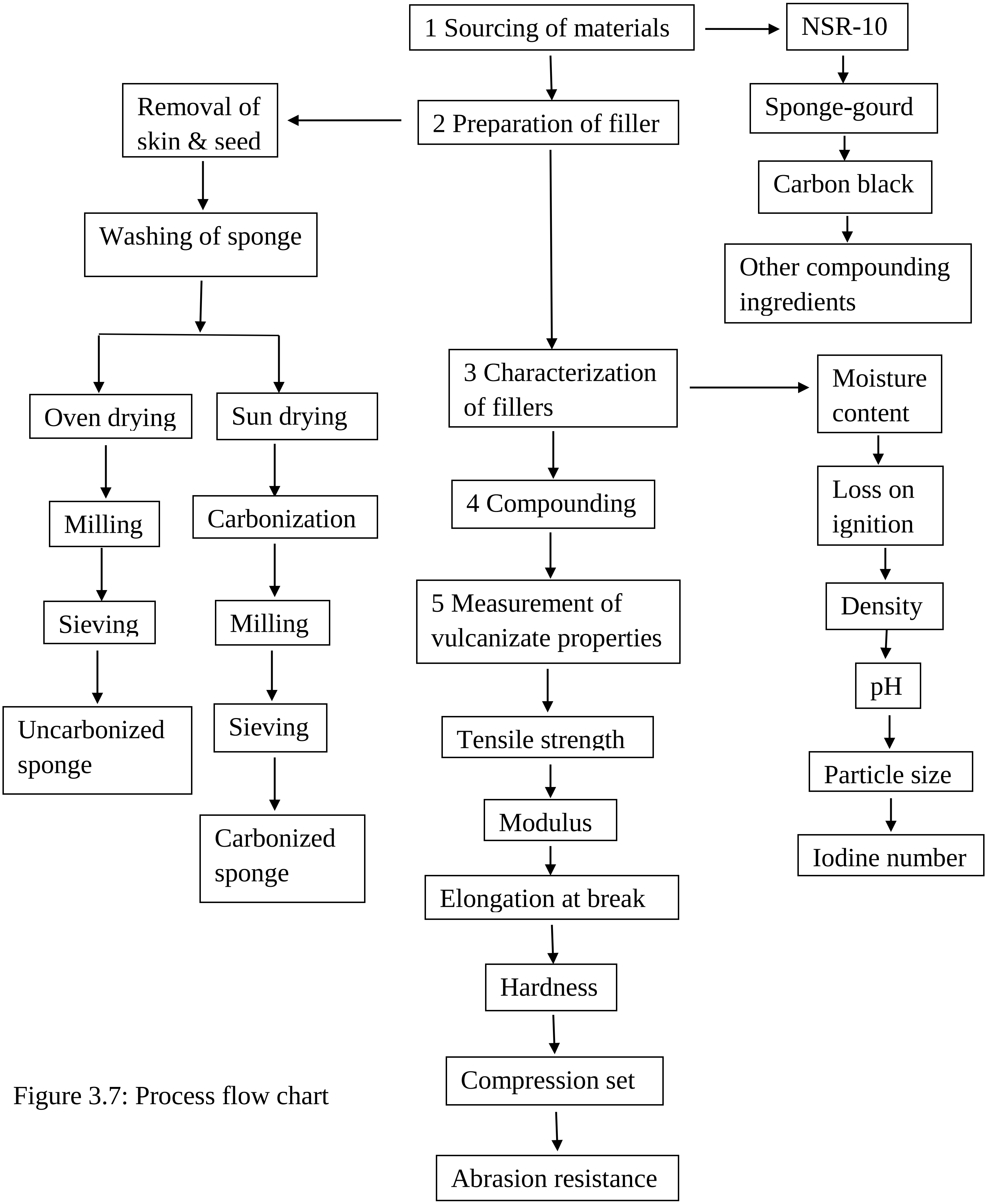

Figure 3.7: Process flow chart

CHAPTER FOUR

RESULTS AND DISCUSSION

4.1 Results

Table 4.1: Physical Characteristics of Fillers

Parameter	US	CS	CB
Moisture Content (%)	11.30	11.40	2.40
Loss on ignition (%)	18.95	20.85	92.60
Density (g/cm^3)	0.23	0.22	
pH of slurry	6.63	8.06	6.50
Particle size (μm)	150	150	150
Iodine adsorption number (mg/g)	12.69	13.32	83.0

Key : US – Uncarbonized sponge ; CS – Carbonized sponge ; CB – Carbon Black

Table 4.2: Tensile strength tests (MPa)

Filler loading	CB	CS	US
3.00	20.50	16.10	14.10
5.00	12.30	16.90	18.50
10.00	16.10	8.00	11.30
15.00	26.50	21.90	15.20
20.00	18.20	16.20	11.70

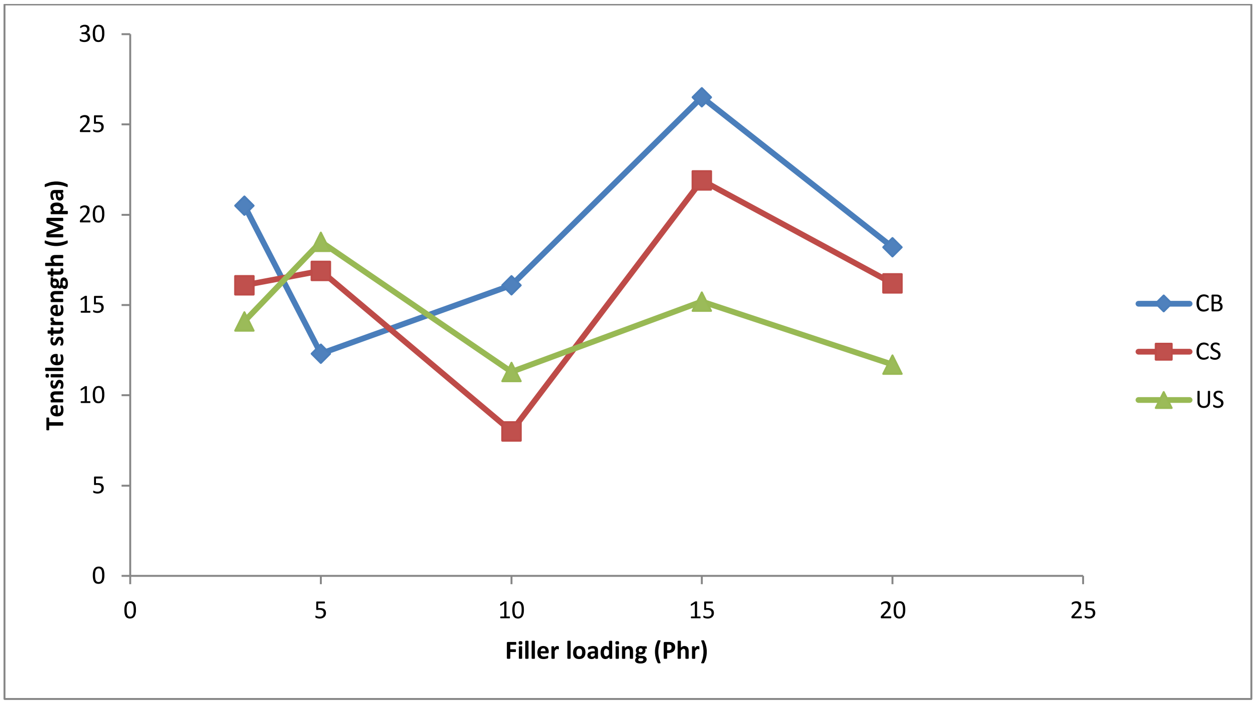

Figure 4.1: Plot of tensile strength versus Filler loading

Table 4.3: Modulus tests (MPa)

Filler loading	CB	CS	US
3.00	0.60	0.50	0.50
5.00	0.50	0.50	0.60
10.00	0.50	0.90	0.80
15.00	0.90	0.60	0.50
20.00	1.20	0.80	0.80

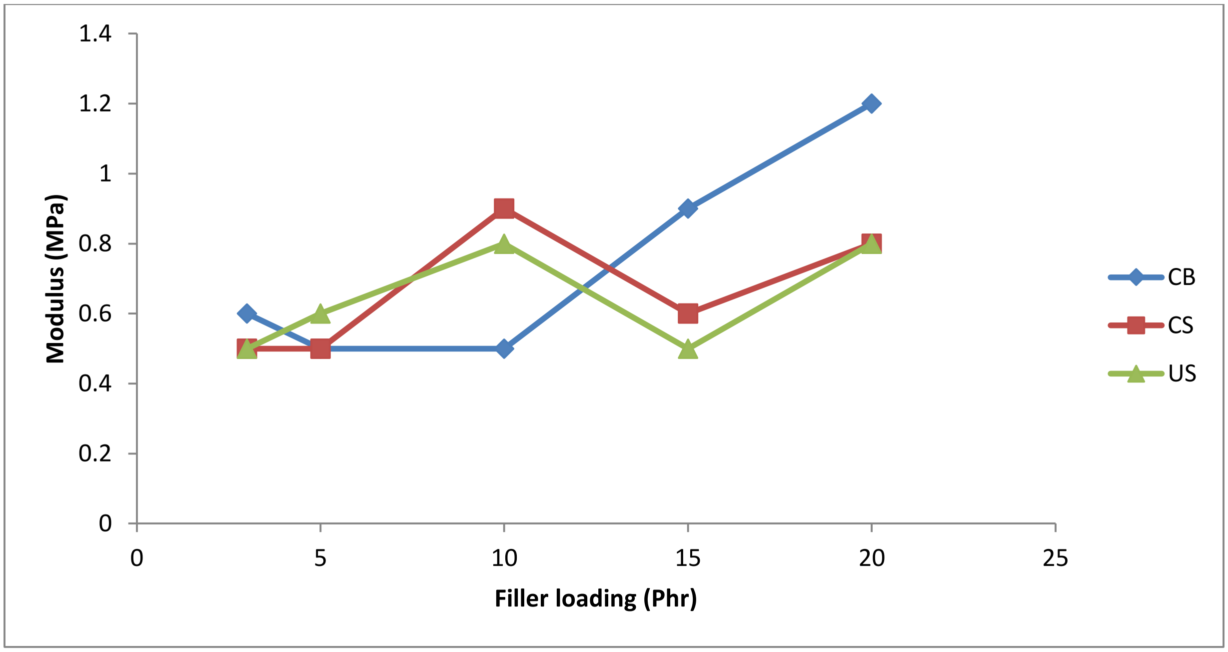

Figure 4.2: Plot of Modulus versus Filler loading

Table 4.4: Elongation at break tests (%)

Filler loading	CB	CS	US
3.00	1196.00	1034.00	1346.00
5.00	1013.00	1129.00	1358.00
10.00	1241.00	675.00	1107.00
15.00	1068.00	1077.00	1472.00
20.00	898.00	965.00	1265.00

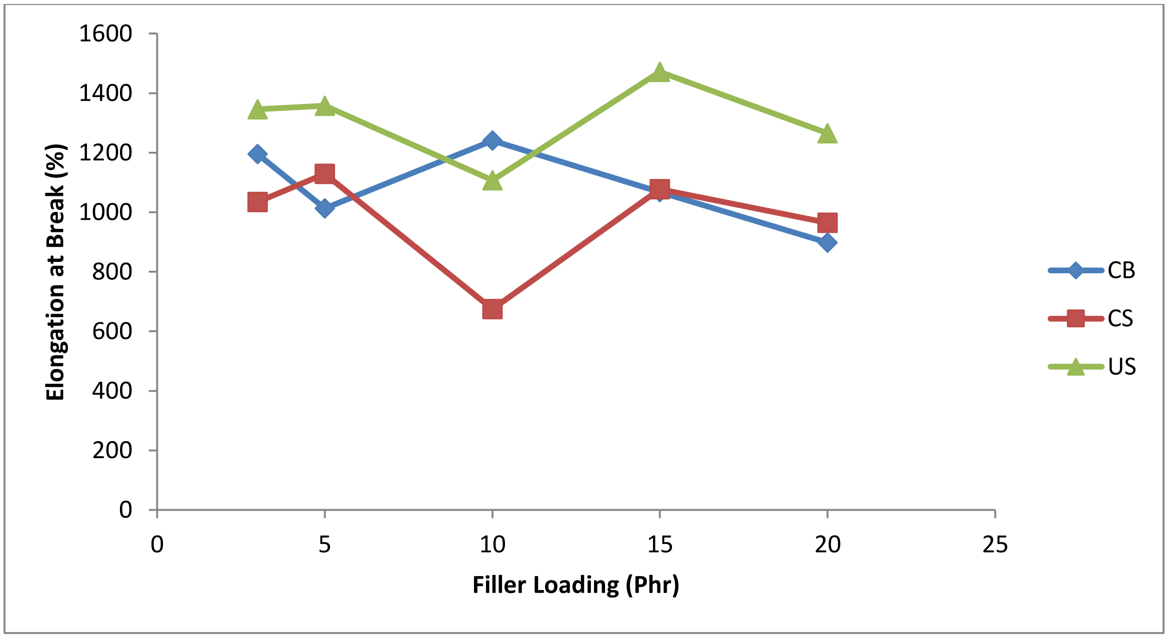

Figure 4.3: Effect of filler loading on EAB of rubber composites

Table 4.5: Hardness tests (IRHD)

Filler loading	CB	CS	US
3.00	43.00	45.00	38.00
5.00	43.00	45.00	40.00
10.00	45.00	47.00	43.00
15.00	49.00	47.00	47.00
20.00	51.00	48.00	55.00

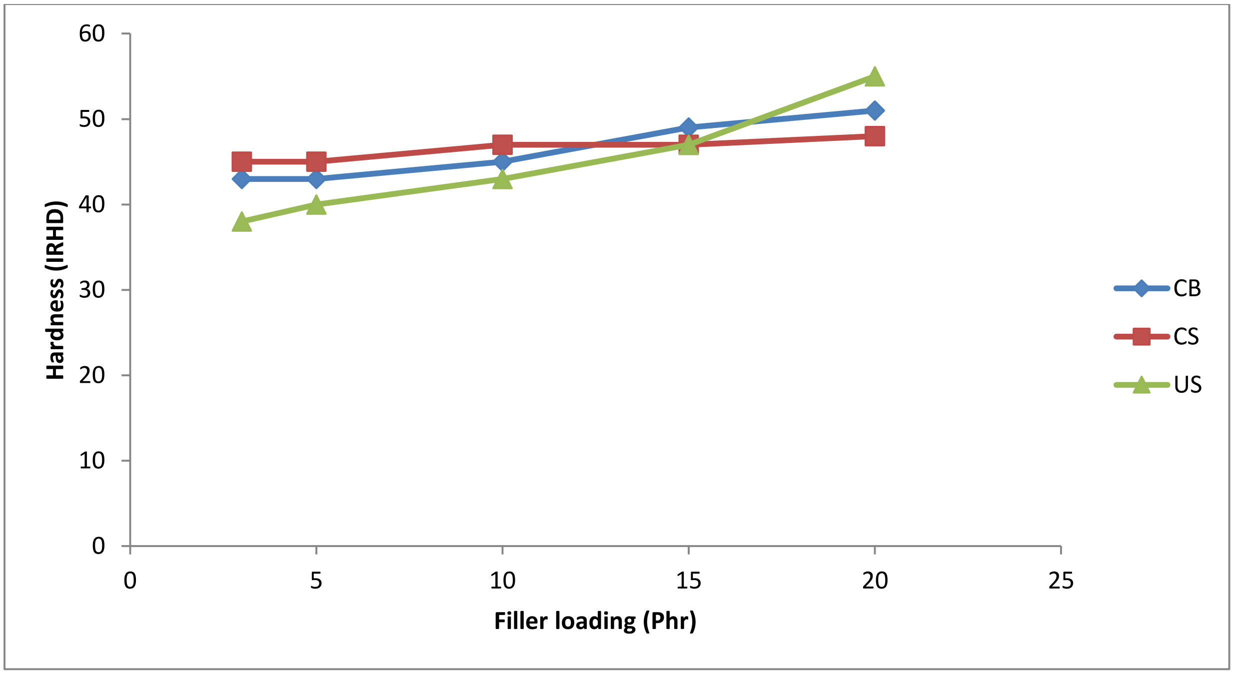

Figure 4.4: Effect of filler loading on hardness of rubber composite

Table 4.6: Compression set test (%)

Filler loading	CB	CS	US
3.00	13.20	12.90	6.15
5.00	11.38	10.07	4.55
10.00	11.00	9.77	2.02
15.00	9.11	10.26	1.75
20.00	8.95	8.25	1.32

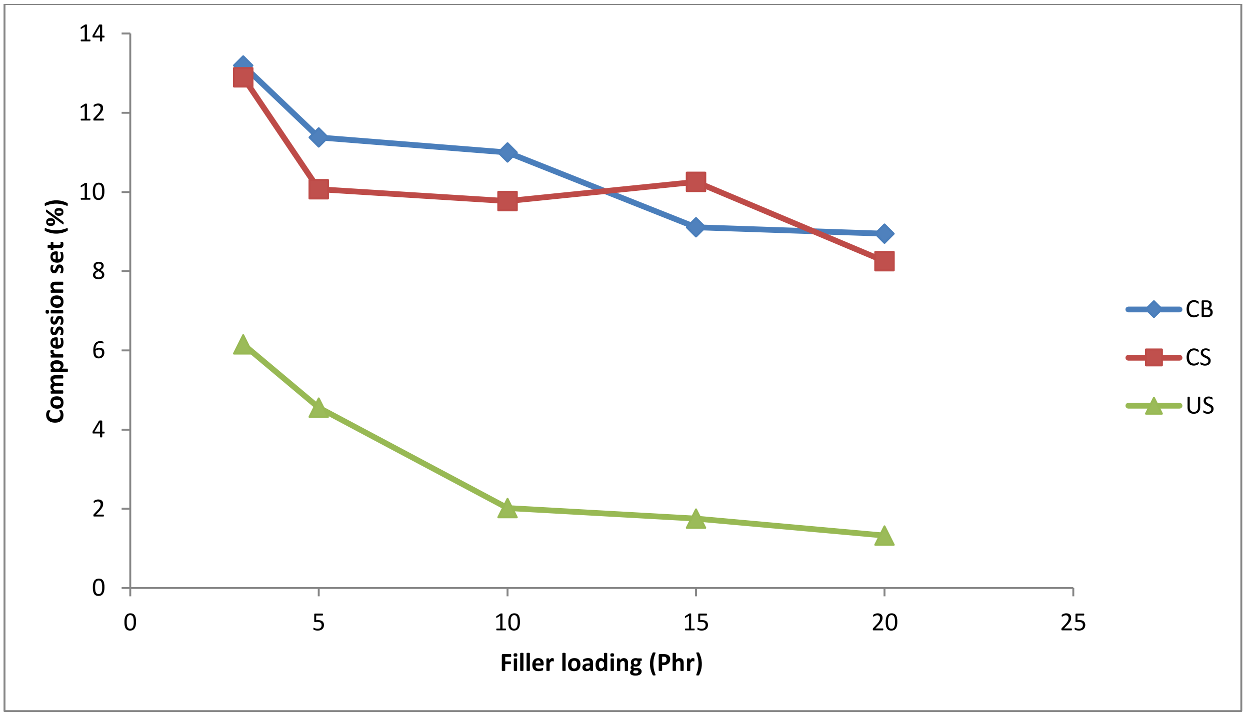

Figure 4.5: Effect of filler loading on compression set of rubber composites

Table 4.7: Abrasion resistance index test

Filler Loading	CB	CS	US
3.00	41.00	60.00	54.00
5.00	37.00	58.00	53.00
10.00	40.00	56.00	54.00
15.00	35.00	55.00	58.00
20.00	37.00	46.00	58.00

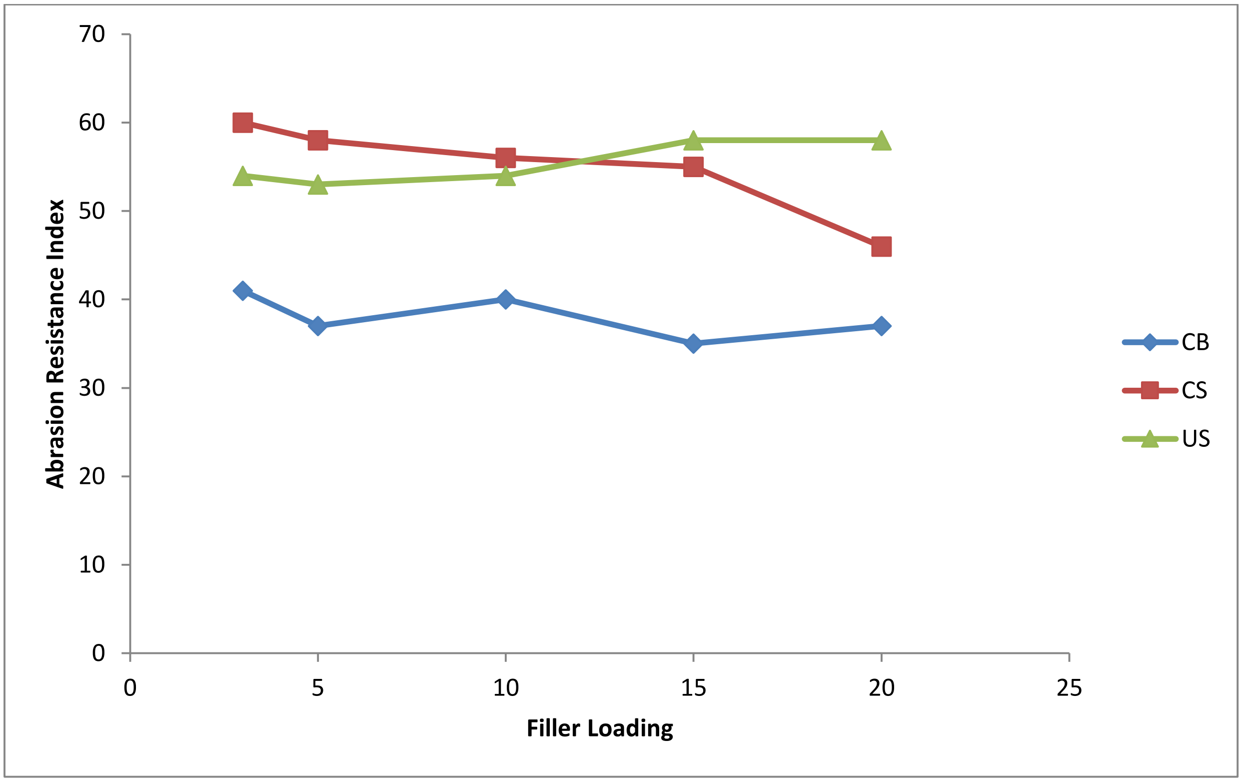

Figure 4.6: Effect of Filler Loading on Abrasion Resistance Index

4.2 Discussion

4.2.1 Physical characteristics of fillers

The result for the characterization of the fillers is as presented in Table 4.1.

Sponge-gourd, being one of the cellulosic fibres, is hydrophilic and absorbs moisture. The result in the above table shows that moisture content for CS and US are 11.40 and 11.30 respectively. It is interesting to note that the moisture content for CS is higher than that of US. This may be attributed to the carbonization temperature. At carbonization temperature of 500℃, the moisture content was still high as reflected by the result. This can lead to dimensional variations in composites and also affects the mechanical properties of the composites (Saheb and Jog, 1999). Besides, during processing, the moisture content can lead to poor processability. In terms of Loss on Ignition, the result show that the value for that of CB is highest and was followed by that of CS. Loss on ignition is the amount of carbon content lost for a given material. It is also a measure of weight loss. This parameter affects the level of distribution and dispersion of fillers in the rubber composites. Higher value implies higher reinforcement potential. This is as observed in this work, as CB filled vulcanizate had the highest reinforcing potential. This was closely followed by CS. On pH it is seen that CS value is higher than that of US. In other words, carbonization raised the pH of the filler from acid for US to alkaline for CS. Review of relevant literature revealed that alkaline fillers increase the cure rates of vulcanizates. For iodine adsorption number, a pointer to the surface area of the filler, the result shows that carbonization raised the iodine number from 12.69 for US to 13.32 for CS. Again higher value means higher reinforcement potential. This was as observed in this study, as CB filled vulcanizates exhibited better reinforcement potential and was followed by CS filled vulcanizates.

4.2.2 Effect of filler loading on Tensile Strength of rubber composites

The tensile strength of composites was studied as a function of filler loading. The results are presented in Table 4.2 and Figure 4.1. The tensile strength of the different samples showed irregular pattern of increase with increase in filler loading. For CB filled vulcanizates, initial decrease at a filler loading of 3Phr was observed. This may be due to the fact that filler loading of 3Phr is lower than the critical fibre volume fraction (Derringer, 1971). The result revealed that for CB the critical fibre volume is 5Phr since it is at this filler loading that the observed decrease stopped and then an increase. Onyeagoro (2010) holds the same view. In his opinion at such a low filler loading, the fillers can but only act as flaws instead of reinforcement. On the contrary, instead of initial decrease, a slight increase was observed for both CS and US filled vulcanizates at a loading of 3Phr. This increase may be that of NR itself, since at such low loading, both fillers did not exhibit reinforcement potential. From the result, it is observed that the reinforcement potential of both CS and US started manifesting at a filler loading of 5Phr, following the initial decrease in tensile strength. The same reasons hold as in the case of CB. It could be seen from the result that CB demonstrated higher reinforcement potential than those of CS and US. The results further show that for both CB and CS, maximum tensile strength was obtained at a filler loading of 15Phr. The sudden decrease in tensile strength above filler loading of 15Phr could be attributed to high fibre population leading to fibre agglomeration. The aftermath of this is difficulty in stress transmission from matrix to fibre and the disturbance of the continuity of the matrix phase. Besides, the decrease in tensile strength, indicate the loading at which the filler bears more of the stress. This is somehow a pointer to the limiting loading levels. A similar observation was reported by Mathew and Joseph (2007) and Arayapranee and Rempel (2008). Generally, the result showed that CB filled vulcanizate showed the highest degree of reinforcement. This was followed by CS filled vulcanizate, while US filled vulcanizate showed the least degree of reinforcement potential. This may not be unconnected to the measured physical characteristics of the fillers as presented in Table 4.1. The results show that weight loss on ignition were 92.60%, 20.85% and 18.95% for CB, CS and US respectively. In physical characterization of fillers, weight loss on ignition is a measure of the carbon content lost during combustion.

It measures the effectiveness of the filler as reinforcement. Onyeagoro (2012) pointed out that the higher the values, the greater the reinforcing potential. The result in Table 4.1 shows an increasing order of reinforcement, which confirms the tensile strength result obtained. Besides, the iodine adsorption number is used to evaluate the surface area of the filler. Again, the values obtained in Table 4.1 indicate that the surface area of CS is significantly higher than that of US, due to carbonization, which improved the reinforcing properties. It is worthy to note that the iodine number obtained for CS may have been affected by the carbonization temperature used in this work. The review of literature pointed out that iodine number increase with increase in activation temperature (Oyoh and Igbokwe, 2001; Ajayi and Olawale 2009 and Yusufu et al, 2012). Another factor that could be responsible for the better reinforcing potential of CS is pH. Table 4.1 also showed that the pH value for CS is higher than that of US. This is as a result of carbonization. It has been reported by Horn (1971), Kumar and Gupta (1998) and Asore (2000) that alkaline fillers increase the cure rates of vulcanizates, as observed during this work.

4.2.3 Effect of Filler Loading on Modulus of rubber composites

The effect of fillers on the modulus, an indication of material stiffness, at 100% elongation is shown in Table 4.3 and Figure 4.2 respectively. Again as in the case for tensile strength, an irregular trend in modulus was observed. This again could be attributed to poor fibre volume fraction. The properties of composites like tensile strength and modulus is strongly influenced by its composition. Regardless of this, the result shows that CB filled vulcanizate had the highest modulus. This was closely followed by CS filled vulcanizate. The moderate moduli may also be due to the particle size of the fillers which is a pointer to their surface area. For good filler – matrix interaction, the surface area of the fillers must be large, which means that the particle size must be small. The observed slight increase at some filler loading, can be explained by the fact that adhesion occurred between the filler and the rubber matrix. This brought about increase in stiffness, rigidity and modulus. More so, the result is a pointer to the fact that modulus of filled vulcanizates depends largely on the level of filler dispersion in the polymer matrix in question. This is in conformity with reviewed literature. But good dispersion is a function of filler properties. This is on the basis that fillers from agricultural waste are usually prone to filler agglomeration. This hinders the dispersion and distribution of the filler in the matrix. Little wonder the report by Wagner (1976) that improving the surface properties of fillers such as surface area, surface reactivity, filler dispersion and filler rubber interaction could enhance the modulus of filled rubber vulcanizates. Kohjiya and Ikeda (2000) hold the same view. The better reinforcing potential of CS when compared to that of US could be attributed to carbonization. The results point to the fact that CS and US will exhibit greater reinforcement if appropriate surface modification is done.

4.2.4: Effect of filler loading on elongation at break (%) of rubber composites

The effect of filler loading on elongation at break is shown in Table 4.4 and Figure 4.3. On Elongation at break, the same irregular increasing pattern was observed when the NR vulcanisate was filled with different fillers. The result indicates that the elongation at break seemed to be affected by low filler loading, surface area of the fillers and also probably by a poor rubber – filler interaction. At similar filler loading of 20Phr, US gave the highest elongation at break value. This was followed by CS and CB with the lowest value. In general, the incorporation of more filler into the rubber matrix results in decrease in elongation at break (Imanah et al, 2003). The decrease with increase in filler loading could be attributed to increase in stiffness and brittleness as a result of the adherence of the fillers to the polymer. The result is resistance to stretching on application of load. This result is in harmony with observations made by other researchers like Ishak and Baker (1995) and Egwakhide et al. (2007).

4.2.5: Effect of filler loading on Hardness of rubber composites.

The hardness values of the NR vulcanizates are shown in Table 4.5 and Figure 4.4. From the result, it is observed that hardness increased with increase in filler loading. The test results for CS and US revealed that they compared excellently well with that of CB which serves as control in this study. Hardness measures small distortion at the surface of the vulcanized elastomer, and is therefore an appropriate indicator for stiffness. It is therefore expected that prolong shelf service life will be exhibited by products made with carbonized and uncarbonized sponge-gourd. The observed increased in hardness was expected. This is because as more filler loadings are introduced into the rubber matrix, the composite becomes more rigid. This is due to the gradual reduction in the elastic property of the rubber. Similar results of increase in hardness with increase in filler loading have been reported by other researchers like Aguele and Madufor (2012), Onyeagoro (2012) and Osabohien et al. (2015).

4.2.6: Effect of filler loading on compression set of rubber composites

When an elastomer is compressed and allowed to relax, it does not return to its initial dimension. This disparity is refered to as compression set. The various rubber composites samples were put through standard compressive loads for a fixed period of 24 hours at 45℃. The extent by which the test piece returns to its primary thickness was evaluated and recorded as compression set. The results of compression set in Table 4.6 and Figure 4.5 show that as filler loading increases, the compression set of filled rubber vulcanizates decreases. According to Sukru et al. (2008), this observation is connected with the degree of filler dispersion and its particle size which may have enhanced its performance. This is expected because the introduction of more filler into the rubber matrix makes it more rigid. The aftermath of this is less susceptibility to compression loading. From the result, US had the lowest value followed by CS and then CB. Low values of compression is an indication of good reinforcement ability. In order words, CB has the least compression set. Similar observation was reported by Egwaikhide et al. (2007).

4.2.7: Effect of filler loading on Abrasion Resistance Index of Rubber composite

The abrasion resistance of the NR vulcanisates is shown in Table 4.7 and Figure 4.6. From the results obtained for all the test samples, it is noticed that increasing filler loading did not continuously increase the abrasion resistance index of the composites and as such had insignificant effect on this property. This irregular behaviour may not be unconnected to the fact that as the filler loading is increased, the adhesion strength between the filler and the polymer matrix falls. The observed pattern may also be due to the degree of dispersion of the fillers. Similar observation was reported by Egwaikhide et al. (2007) and Aguele et al. (2014).

CHAPTER FIVE

CONCLUSION AND RECOMMENDATION

5.1 Conclusion

In this study, CS and US were comparatively investigated as fillers in NR composites. The result has shown that CS and US filler influenced the mechanical properties of natural rubber vulcanizates. Though, a general trend for reinforcement was observed for both CS and US as indicated in the results obtained during this study, the reinforcement potential was more for carbonized (CS) than uncarbonized (US) fillers. This is a pointer that the vulcanizate properties of sponge-gourd powder filled natural rubber composites can be improved by carbonization. Besides, it reveals that CS can serve as a potential substitute filler for CB, especially for the production of low cost/high volume rubber products were strength is not critical. Finally, the result show that properties tested were appreciably affected by filler loading .The results for tensile strength, modulus, elongation at break and abrasion resistance show that CB had the highest reinforcing potential. This was closely followed by CS. The fact that sponge-gourd filler imparted lower reinforcing effects for the properties mentioned than the commercial grade carbon black, may be due to larger particle size, small surface area and small loss on ignition when compared to that of carbon black.

5.2 Recommendation

It is pertinent to state that there are other related areas of study that could benefit this work in sponge-gourd reinforced NR composites. For example, further research could focus on areas identified below.

Carbonization could be done at various temperatures and time, so as to ascertain the best carbonization temperature and time duration for sponge-gourd. This could have reasonable effects on the final reinforcement imparted on rubber composites.

Furthermore, better distribution of filler volume like 10, 20 30, 40 and 50Phr should be used. This will allow a clearly defined trend to be obtained from the results.

More so, modification may be done on the filler by way of alkali treatment, use of compatibiliser and/or coupling agent. This could result in better fibre-matrix adhesion and further improvement in the reinforcement potentials of the fillers.

Finally, Scanning Electron Microscopy (SEM) could be carried out on sponge-gourd powder filled natural rubber composites. This will validate the improvement in mechanical properties reported in this work.

Contribution to knowledge

To the best of my knowledge, the work by other researchers in the polymer industry have concentrated on the use of sponge-gourd in the reinforcement of other polymers like epoxy resins, polyester resins, resorcinol formaldehyde etc. But this study has opened a new area of research in this field, using sponge-gourd in the reinforcement of natural rubber.

Besides, this study has added to the number of available literature on sponge-gourd. This is important following the lamentation by other researchers for inadequate research on sponge-gourd fibre.

Finally, the knowledge that sponge-gourd can be used for the reinforcement of NR will result in its use as alternative to carbon black especially in the production of low cost/ high volume rubber products were strength is not critical.

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