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Tensile Properties of Treated and Untreated Ground Nut Shell-Filled Natural Rubber Composites

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Abstract: *Groundnut shell was crushed in to particle size and given two surface treatments with alkali and 3-chloro-2-hydroxylpropyltrimethylammoniumchloride respectively. The raw, alkali-treated and bonding agent treated groundnut shell cellulose fibers were used as natural rubber composites. The samples were used to produce fiber-reinforced natural rubber composite at varying filler loadings. Properties such as tensile, hardness and impact of the composites were investigated. The tensile strength of the composites varied such that both the alkali-treated and cationized fillers recorded higher values than untreated fillers. The impact hardness properties were also found to be better in the modified fillers than the untreated ones. This work has shown some general improvements arising from cauticization and cationization of cellulosic filler as reinforcing material for natural rubber.*
Key words: *Cationization, Custicization, Ground nut shell, Natural rubber, Tensile properties*

I. Introduction

Natural rubber also has been called ‘the supreme agricultural colonist of all times’. It is originally indigenous from the Amazon Valley forest, but has been cultivated principally in Southeast Asia, especially for countries like Malaysia and Indonesia. Where more than fourteen million acres of land have been cleared and planted with rubber trees. A considerable amount of research has been done in the field of fiber reinforced elastomer composites. Researchers have studied the effect of different fibers in natural and synthetic rubber for most of these researches petroleum-based resources have been used however, they are non-biodegradable and their disposal contributes to many environmental problems and this prompt researchers to develop biodegradable materials [1]. The investigation of physic-mechanical properties carried out on natural rubber-coconut fiber composites, showed that coconut fiber is potential reinforcing filler for natural rubber compounds. In addition, palm kernel husk was also found to be potential reinforcing filler for natural rubber compounds [2]. Previous work also indicated that the use of lignocellulosic fibers as fillers can improve the properties of polymers and this research work intends to explore the possibilities of using groundnut shell cellulose fibers as potential reinforcement in natural rubber. The most important parameters that affect the fiber-reinforcement are fiber loading, fiber dispersion, fiber orientation and adhesion between the fiber and the matrix [3].

Natural cellulose fiber has the potential to be an attractive alternative to synthetic fiber and is currently being explored in sectors such as the automobile and building industries. In addition, these fibers offer an excellent for use and are of abundant sources. Natural fibers have advantages over synthetic fibers because of their renewable nature, low cost, biodegradability and ease of chemical modification [4].

II. Experimental

The reagents used were prepared using standard analytical methods of preparation. The cationizing agent (3-chloro-2-hydroxylpropyltrimethylammoniumchloride) used was obtained from Sigma Aldrich and used as received.

2.1 Materials

Crumb natural rubber which conforms to technically specified rubber (TRS) was obtained from Rubber Research Institute of Nigeria (RRIN), Ayonomo, Benin City Edo State. The groundnut shell was obtained from Madobi, Madobi Local Government Area, Kano State.

2.2 Sample preparation

The samples were prepared by grinding using a cleaned and dried grinding machine followed by sieving to the particle size of 1mm.

2.3 Moisture content determination

1g of each sample was put in a watch glass and kept at room temperature for 48hours. The watch glass containing the sample was recorded as initial weight (W_1). They were placed in an ovum and maintained the temperature of 100^0C and continue to weigh at regular intervals of 30minutes till a constant weight was recorded. The weight was recorded as the final weight (W_2). The moisture content was determined using the relation:

$$\% \text{ Moisture} = \frac{W_1 - W_2}{W_1} \text{ x } 100$$

Where W_1 = initial weight, and W = final weight [5].

2.4 Causticization (Alkali treatment)

500g of 1mm size sample was cautisized by the use of 20% (owf) sodiumhydroxide solution for 24hours at room temperature after dilution with 7 liters of distilled water. The wet fibers were squeezed to remove excess water and then washed repeatedly to remove excess NaOH. It was finally dried in an ovum at 80^0C for 5hours, and pounded to 120µm particle size [5].

2.5 Cationization

500g of the caustisized fiber were cationized by the bonding agent 65% (3-chloro-2-hydroxylpropyltrimethylammoniumchloride) at 10% owf in the presence of 10.92% NaOH after dilution with 7800ml distilled water. The mixture was kept at room temperature for 24hours maintaining the pH of 12 by periodic addition of the 10.92% NaOH and agitation. The cationized fiber was washed repeatedly to remove the excess NaOH and the bonding agent. The wet fibers were put in an ovum at 80^0C for 5hours to dry and pounded to 120µm particle size [5].

2.6 Compounding

The recipe used in the formulation of the natural rubber composites is given in the table below. Mixing was carried out on a laboratory two-roll-mill in accordance with the method described in the American Society for Testing and Materials (ASTM-D3184-80). Cured samples produced on electrically heated press at 150^0C for 40minutes under a pressure of 7torrs.

Table 1: Typical formulation of rubber compounds

Ingredients	Quantity (phr)
Natural rubber	100
Filler	0-50
Zinc oxide	5.0
Stearic acid	1.5
Sulphur	2.5
MBTS	0.5

MBTS = Mercaptobenzthiazoledisulfide

2.7 Determination of Physico-mecanical Properties

2.7.1 Determination of Tensile Strength

Tensile properties of the cured composites were measured with Instron universal tester at cross-head speed of 60mmmin^{-1}using dumb bell shaped test specimens as contained in ASTM-D412-87

2.7.2 Impact Test

The impact test was carried out using Resil compactor, model 16650 type 6957 in accordance with the British standard BSEN 6603-2. The total impact mass used is 1.00kg.

2.7.3 Hardness Test

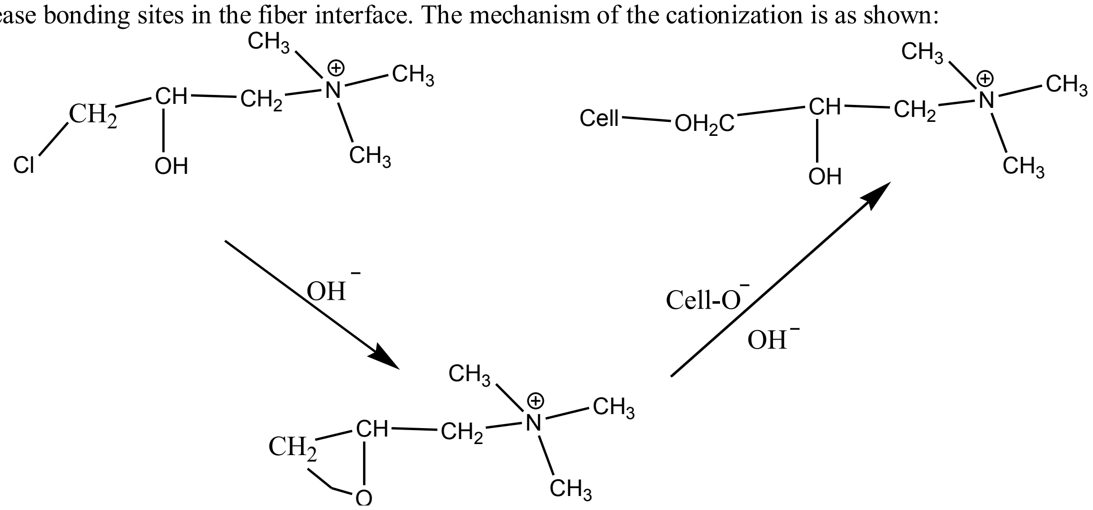
Hardness of the cured composites was measured in shore A, by Durometer instrument, model 5019. The measurement was in accordance with ASTM-D2240.

2.8 Fourier Transform Infra red Spectroscopy (FTIR)

FTIIR spectra of the compounded Groundnut shell-Natural rubber were recorded on 8400s Fourier transform infrared spectrophotometer using KBr pellet technique in the range 4500 – 400cm^{-1} with a resolution of 2cm -^{1}.

III. Results and Discussion

The alkali treatment (causticization) removed pectin and other soluble carbohydrates like hemi-cellulose, leaving behind the alkali resistance cellulose. This exposes the hydroxyl group of the fiber and increase bonding sites in the fiber interface. The mechanism of the cationization is as shown:



The moisture content of the dry fiber, and cationized fibers show the values of about 8%, 7.2% and 5.6% respectively. This gives a good dispersion of the fibers in the natural rubber matrix. Similar report was given by [6].

Broad absorption band of hydroxyl group around 3790 – 3169 cm^{-1} is attributed to the OH stretching vibrations of cellulose, hemicelluloses and water absorbed constituents of the fiber. The OH stretching decreased with custicization, and cationization. The C=O stretching vibration of carbonyl groups in hemicelluloses in the fiber can be seen at peak near 1733 cm^{-1}. Those of the custicized and cationized samples have been shifted to 1603cm^{-1} and 1722cm^{-1} respectively. The presence of aromatic ring in the sample can be seen at 1600 – 1400cm^{-1}. The absorption band near 1400 – 1300cm^{-1} may be attributed to aliphatic C-H in the plane deformation vibration of methyl or methylene groups. The C-O stretching vibration of aliphatic primary and secondary alcohols in cellulose can be seen in band region 1300 – 1000 cm-1 [1].

3.1 Tensile Strength

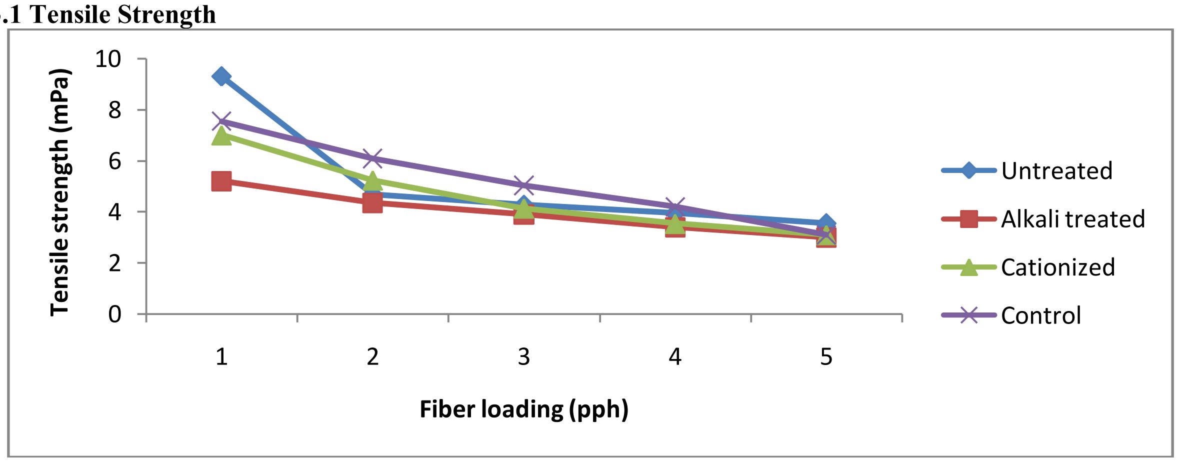


Figure 1: Effect of fiber loading on tensile strength of natural rubber-groundnut shell composites

The tensile strength of the composites shown in fig. 1 and 2 indicated strength decrease with increase in fiber loading. The rubber molecules are themselves internally cross-linked, possessing rather high tensile strength as a result of the so called strain-induced crystallization [5]. However, the addition of the fiber interfered with this natural tendency leading to disruption of the regular arrangement of the molecules, resulting in the loss of ability to crystallize and hence the observed decrease in tensile strength with increase in fiber loading. Figure 1 showed that the tensile strength of the composite filled with causticized and cationized fibers were higher compared to composites made with untreated fibers at most loadings especially of 20 to 40 phr. From the result, it is clear that the alkali and bonding agent treated cellulosic fibers gave higher adhesion to rubber matrix [7]. The effect was more pronounced in the cationized fiber composites.

3.2 Impact Properties

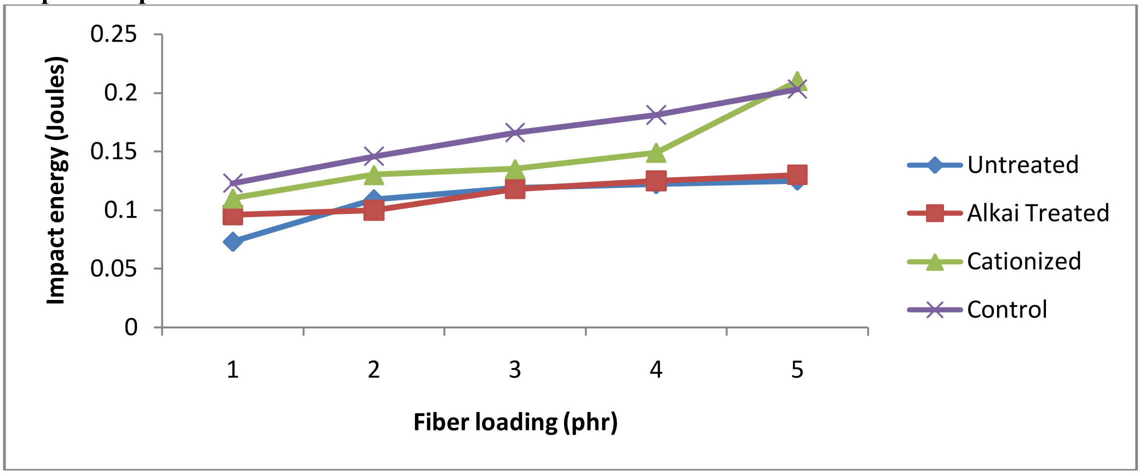


Figure 2: Effect of fiber loading on impact strength of natural rubber-groundnut shell composites

From fig. 2 it can be observed that cationized fibers show better impact properties than the alkali-treated and untreated fibers. The insensitivity of surface treatment with alkali on impact strength was reported by Chuai and his co-workers [8]. Generally, cationized fibers show better impact properties than the alkali-treated and untreated fibers.

3.3 Hardness Properties

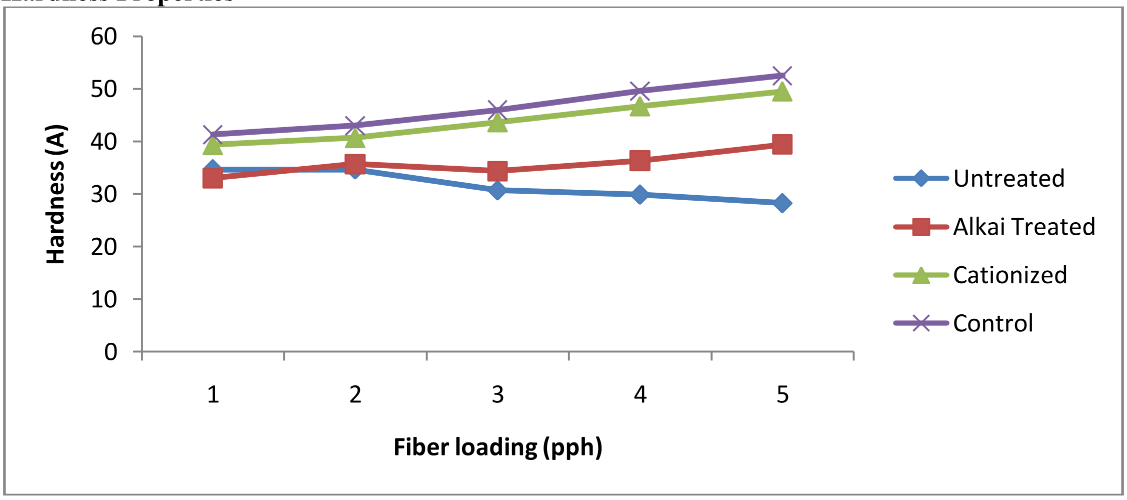


Figure 3: Effect of fiber loading on hardness of natural rubber-groundnut shell composites

The hardness results in fig. 3 of the different samples increased with increasing fiber content. This is in line with the reduced elasticity as a result of the reinforcement in the rubber molecules. Yakubu et. al [5], credited this increase in hardness with fiber loading to the increase in rigidity as the elasticity of the virgin rubber decreases. In the case of untreated groundnut shell fiber, hardness increased with increase in fiber content of the composite of up to 20% weight fraction. This is because there is decrease in coalescence of the fiber particles which also reduces the rate of getting in of the fiber in to rubber, hence reduced the hardness property [9, 10]. In each curve, the cationized fiber-reinforced sample gave higher hardness ratings while causticized sample is intermediate between the untreated and cationized samples.

IV. Conclusion

Two different treatments (custicization and cationization were carried out on ground nut shell cellulosic fiber and the treated as well as the untreated fibers used for the compounding of natural rubber. The Mechanical properties such as impact strength and hardness of both fibers were found to increase with increase in fiber loading. While tensile strength decreases with increase in fiber content of the composites. Therefore the use of wood flour or groundnut shell fibers as filler for natural rubber is of economical value and their consumption as filler would help in reducing environmental waste.

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