

## Utilization of carbonized palm kernel shell as filler in natural rubber composite

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### ABSTRACT

Utilization of carbonized palm kernel shell filled natural rubber compound was studied. Samples of the palm kernel shells were carbonized at varying temperatures (100, 200, 300, 400, 500, 600 and 700°C) for 3 hours each and sieved through a 100 $\mu$ m and 150 $\mu$ m mesh respectively. The characterization results show that pH, iodine adsorption number and loss on ignition increase with carbonization temperature, moisture content decreases, unlike bulk density which decreases and increases with carbonization temperature. The mechanical properties of the 100 $\mu$ m carbonized palm kernel shell filled vulcanizates present better tensile strength, modulus at 100%, abrasion resistance, compression set, rebound resilience and hardness as carbonization temperature increases from 100 - 600°C and starts decreasing as the temperature increases >600°C while the 150 $\mu$ m filled vulcanizates presents better Elongation at break and flex fatigue resistance.

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### KEYWORDS

Carbonization;  
Vulcanizate;  
Palm-kernel;  
Characterization resilience;  
 $\mu$ m.

### INTRODUCTION

The demand for polymers in many applications has experienced a steady growth over the years. In developed countries of Europe and America, the volume of polymers used is more than metals or ceramics<sup>[1]</sup>. Rubbers, being a major form of polymers are not left out. Rubber in its raw state, have virtually no engineering application, therefore, it is necessary to appropriately compound them in order to obtain good processing characteristics, and the desire cure properties in the finished products<sup>[2]</sup>. It is not surprising, therefore, that more than 50% of all chemists and chemical engineers, large numbers of physicists and mechanical engineers and nearly all

material scientist and textile technologists are involved with research or development work with polymers<sup>[3]</sup>. Rubbers as one of the classes of polymers have found great value in many applications including engineering, sport and leisure, and domestic. In its raw state, rubber may not be good enough for any useful application so there is the need for the addition of additives which help to enhance the properties<sup>[4]</sup>. Among these additives are fillers. Carbon black is the major filler used in rubber industry. Today, there are a lot of research going on to find an alternative (local source) for it. Advances are being made into the use of other agricultural by-products, such as rice husk, groundnut shell, rubber seed shell etc.<sup>[5]</sup>. These materials are used in their raw form or

## Full Paper

they are modified before use. The importance of fillers in rubber production cannot be over-emphasized. It is the most widely used in terms of volume in the rubber industry. The most commonly used fillers are the carbon black, china clay, calcium carbonate etc. Of all these fillers, carbon black is the most used for rubber product except where colour is of priority<sup>[6]</sup>. The importation of carbon black is very expensive. It is also a product of petroleum, a source which is non-renewable. Therefore, the need to source for alternative material from local source with little modification becomes necessary. Thus this research is aimed at studying the effect of carbonization temperature on the filler characteristics and on the mechanical properties of the composites.

### EXPERIMENTAL PROCEDURES

#### Materials

#### Equipments used include

- Monsanto Tensile Tester Model 1/m, Manufactured by British Company Limited, England
- Wallace Hardness Tester Model c8007/25, Elektron Technology Series, UK.
- Wallace Akron Abrasion Tester, Elektron Technology Series, UK.
- DuPont Machine, Manufactured by British Company Limited, England
- Muffle Furnace METTm-525, Elektron Technology Series, UK.
- Two Roll Mill, Manufactured by British Company Limited, England
- Hydraulic press, Elektron Technology Series, UK.

#### Method

#### Filler preparation and characterization

Carbonization of fillers used in this research work was carried out using a laboratory muffle furnace set at varying temperatures from 100, 200, 300, 400, 500, 600 and 700°C by creating an inert environment via the passage of nitrogen gas for three hours each<sup>[7]</sup>. The Palm kernel shells were washed in water and dried in air to remove sand particles and moisture respectively. After drying, a portion of the palm kernel shells were milled to fine powder as well as the carbonized portion, and sieved through a mesh size of 100µm and 150µm respectively. The fine particles that passed through were collected and used for compounding. Thus, carbonization is a process of converting an organic compound to high carbon content by heating it in the absence of oxygen (inert).

#### Processing of the composites

The formulation used for the compounding of the natural rubber (NSR-10) with the palm kernel shell fillers is given in TABLE 2

#### Mixing procedure

The rubber mixes were prepared on a laboratory size two roll mill according to the mixing cycle shown in TABLE 3. It was maintained at 70°C to avoid cross-linking during mixing after which the rubber composite was stretched out. Mixing follows ASTM D 3184–80 Standard<sup>[9]</sup>.

#### Compounded composite curing

The curing of test pieces was carried out at 130°C for 10 minutes under a pressure of 2.8bar.

### RESULTS AND DISCUSSION

#### Results

TABLE 1 : Materials and their sources

| Materials                                  | Sources   |
|--|---|
| Natural Rubber (NSR-10)                    | Rubber Research Institute, Iyanomoh, Benin City |
| Palm Kernel Shells                         | Eziobodo, Owerri, Imo State, Nigeria            |
| Tetramethyl thiuram disulphide (TMTD)      | British Drug House (BDH), England               |
| Mercapto benzothiazole sulphenamide (MBTS) | Vulnax International Ltd, Warrington, England   |
| Stearic acid                               | British Drug House (BDH), England               |
| Sulphur                                    | British Drug House (BDH), England               |
| Trimethylquinoline (TMQ)                   | Vulnax International Ltd, Warrington, England   |
| Zinc Oxide                                 | Vulnax International Ltd, Warrington, England   |
| Processing Oil (Paraffin Wax)              | British Drug House (BDH), England               |
| N330 Carbon black                          | Vulnax International Ltd, Warrington, England   |

TABLE 2: Formulation for compounding natural rubber<sup>[8]</sup>.

| Ingredient                    | Parts per hundred rubber |
|-------------------------------|--------------------------|
| Natural rubber                | 100                      |
| Fillers (CPKS)                | 40                       |
| Zinc Oxide                    | 5.0                      |
| Stearic acid                  | 2.5                      |
| Sulphur                       | 1.5                      |
| MBTS                          | 1.5                      |
| TMTD                          | 3.5                      |
| Processing Oil (Paraffin Wax) | 5.0                      |

A batch factor of four (7) was used.

TABLE 3: Mixing steps and time

| Mixing Steps               | Time (minutes) |
|----------------------------|----------------|
| Natural rubber mastication | 6              |
| Addition of Stearic acid   | 1              |
| Addition of Zinc Oxide     | 2              |
| Addition of filler (CPKS)  | 9              |
| Addition of MBTS           | 1              |
| Addition of TMTD           | 1              |
| Processing Oil             | 1              |
| Addition of Sulphur        | 1              |
| Total                      | 21             |

## DISCUSSION

The results of the vulcanizates shown in TABLE 4 presents a gradual increase in tensile strength as well as modulus with carbonization temperature of the filler is noticed up to 600°C. It may be mentioned here that both tensile strength and modulus are important for recommending any vulcanizate for structural applications. The increase in tensile strength and modulus for the 100µm-filled vulcanizates than 150µm-filled vulcanizates is as a result of high surface area, suggesting better polymer filler interaction and hence enhanced better tensile properties. The factors that affect the reinforcing potentials of fillers include filler dispersions, surface area, surface reactivity, bonding capacity (quality), particle size between the filled and the elastomeric matrix<sup>[10]</sup>.

The modulus data in TABLE 4 and Figure 2 showed increase with filler carbonization temperature for the 100µm-filled and 150µm-filled vulcanizates up to 600°C before showing a marked decrease with temperature >600°C. The fact that

100µm-filled has higher modulus than 150µm-filled vulcanizates, suggest that fillers are more reinforcing when properly adhered into the polymer matrix.

The results in TABLE 4 and Figure 3 showed that elongation at break (EAB) decrease with increasing filler carbonization temperature of the mixes for all the fillers below 600°C, above which there was a gradual rise in the value of EAB. A decrease in EAB has been explained in terms of adherence of the filler to the polymer phase leading to the stiffening of the polymer chain and hence resistance to stretch when the strain is applied<sup>[11]</sup>. 100µm-filled vulcanizates has less values of EAB as temperature increases compared to 150µm-filled.

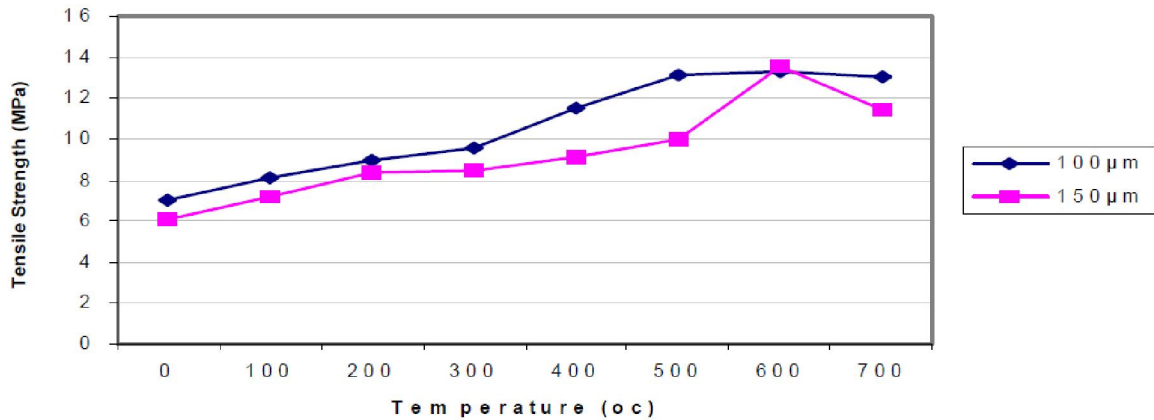
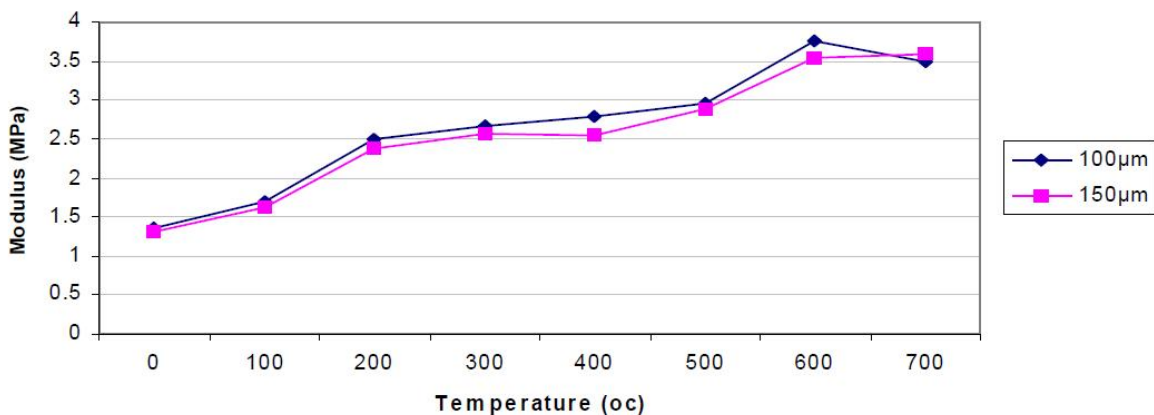
The hardness of 100µm-filled and 150µm-filled vulcanizates increased with increasing carbonization temperature. However, above 600°C the trend took a different direction as a result of the filler characteristics. This result is expected because as more filler particles get into the rubber, the elasticity of the rubber chain is reduced, resulting in more rigid vulcanizates. The hardness results of 100µm-filled vulcanizates are higher than those of 150µm-filled as shown in TABLE 4 and Figure 4 because of the

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**TABLE 4 : Mechanical properties of the carbonized palm kernel filled vulcanizates**

| Properties                         | N330 Carbon black | Uncarbonized         | Carbonization Temperature (°C) at 40phr Filler Loading |                      |                      |                      |                      |                      |                      |
|------------------------------------|-------------------|----------------------|--|----------------------|----------------------|----------------------|----------------------|----------------------|----------------------|
|                                    |                   |                      | 100  | 200                  | 300                  | 400                  | 500                  | 600                  | 700                  |
| Tensile Strength (MPa)             | 29.20             | (7.00)<br>[6.08]     | (8.11)<br>[7.22]                                       | (9.00)<br>[8.35]     | (9.55)<br>[8.47]     | (11.54)<br>[9.16]    | (13.15)<br>[10.01]   | (13.32)<br>[13.56]   | (13.07)<br>[11.45]   |
| Modulus at 100% (MPa)              | 6.30              | (1.35)<br>[1.30]     | (1.70)<br>[1.63]                                       | (2.50)<br>[2.37]     | (2.67)<br>[2.56]     | (2.78)<br>[2.55]     | (2.76)<br>[2.85]     | (3.75)<br>[3.55]     | (3.48)<br>[3.48]     |
| Elongation at break (%)            | 324.10            | (589.04)<br>[600.00] | (568.05)<br>[585.00]                                   | (550.04)<br>[575.21] | (538.78)<br>[550.44] | (523.50)<br>[532.09] | (479.44)<br>[498.05] | (462.54)<br>[425.08] | (399.99)<br>[475.08] |
| Flex fatigue (kcx10 <sup>3</sup> ) | 4.58              | (9.98)<br>[10.02]    | (9.02)<br>[9.70]                                       | (8.45)<br>[8.42]     | (7.44)<br>[7.75]     | (7.21)<br>[6.43]     | (6.00)<br>[6.90]     | (6.00)<br>[6.88]     | (6.87)<br>[7.19]     |
| Hardness (IRHD)                    | 58.60             | (50.09)<br>[50.00]   | (53.06)<br>[52.00]                                     | (54.49)<br>[53.07]   | (60.95)<br>[60.00]   | (61.12)<br>[60.34]   | (64.24)<br>[64.67]   | (68.11)<br>[67.90]   | (67.75)<br>[61.06]   |
| Abrasion Resistance (%)            | 40.60             | (20.09)<br>[20.10]   | (21.00)<br>[20.25]                                     | (22.08)<br>[21.00]   | (34.97)<br>[33.42]   | (29.00)<br>[37.25]   | (49.78)<br>[44.20]   | (51.43)<br>[51.04]   | (52.20)<br>[49.90]   |
| Compression set (%)                | 8.61              | (20.53)<br>[20.02]   | (20.85)<br>[20.18]                                     | (17.71)<br>[17.90]   | (15.08)<br>[15.09]   | (13.32)<br>[13.08]   | (12.91)<br>[11.16]   | (9.31)<br>[9.08]     | (9.15)<br>[9.78]     |
| Rebound Resilience (%)             | 92.67             | (87.70)<br>[87.70]   | (78.60)<br>[77.90]                                     | (76.90)<br>[75.40]   | (73.60)<br>[73.80]   | (87.80)<br>[83.60]   | (82.00)<br>[78.80]   | (76.90)<br>[69.80]   | (86.10)<br>[73.80]   |
| Specific Gravity(SG)               | 1.021             | (1.006)<br>[1.006]   | (1.009)<br>[1.042]                                     | (1.066)<br>[1.081]   | (1.102)<br>[1.108]   | (1.006)<br>[1.016]   | (1.040)<br>[1.050]   | (1.066)<br>[1.083]   | (1.008)<br>[1.019]   |

Key: 100µm-filled vulcanizates ( ), 150µm-filled vulcanizates [ ]


**Figure 1 : Tensile strength of 100µm-filled and 150µm-filled vulcanizates**

**Figure 2 : Modulus at 100% of 100µm-filled and 150µm-filled vulcanizates**

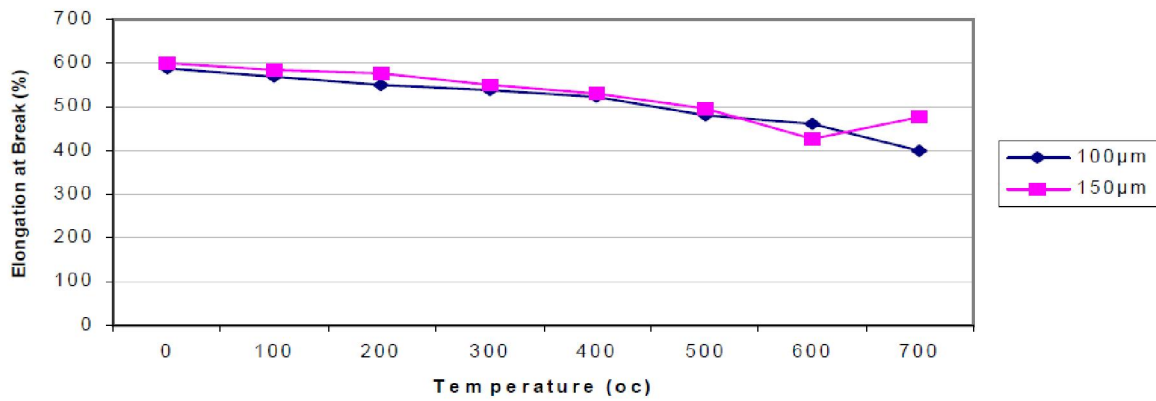


Figure 3 : Elongation at break of 100µm-filled and 150µm-filled vulcanizates

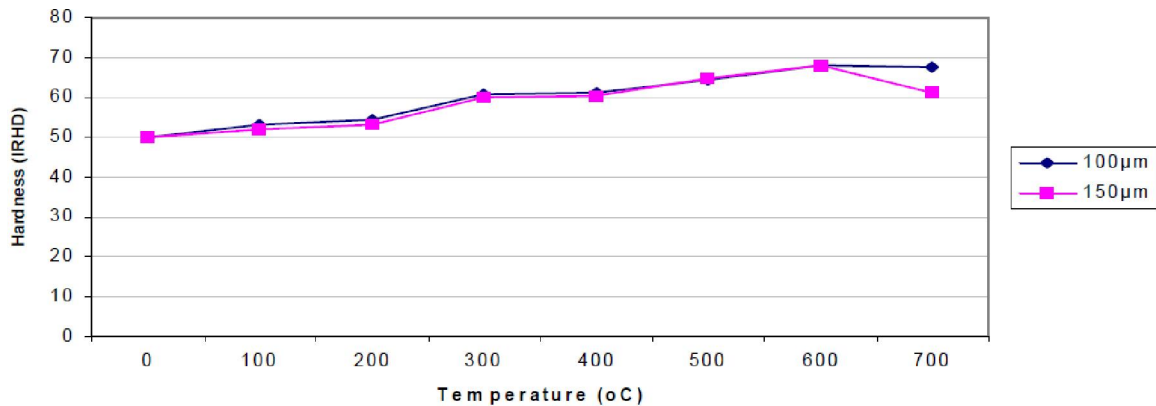


Figure 4 : Hardness of 100µm-filled and 150µm-filled vulcanizates

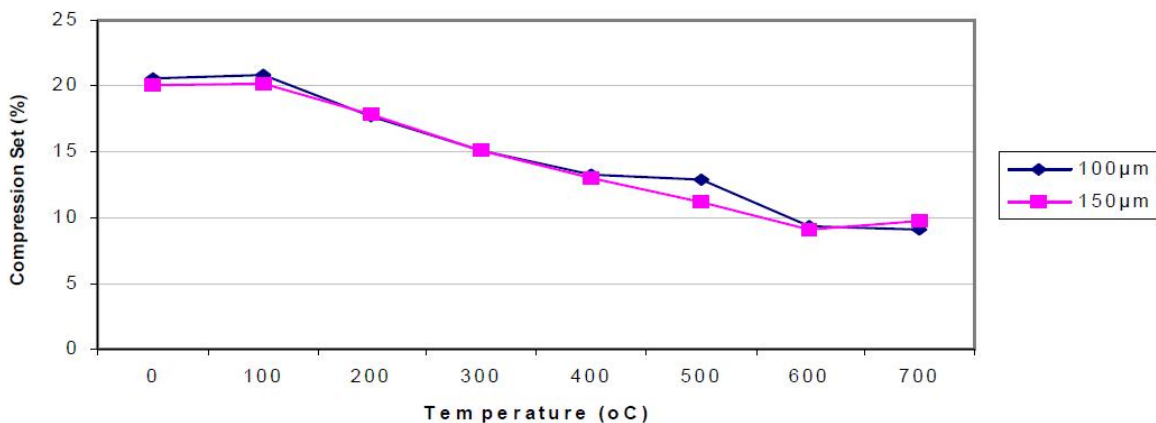


Figure 5 : Compression set of 100µm-filled and 150µm-filled vulcanizates

more adherence of 100µm carbon to the rubber matrix.

The results of compression set in TABLE 4 and Figure 5 show that as filler carbonization temperature increases, the compression of filled vulcanizates decreases for both 100µm-filled and 150µm-filled vulcanizates up to 700°C. This observation is connected with the degree of filler dispersion and its particle size which may have enhanced the 100µm-filled vulcanizates<sup>[12]</sup>. However, the values for both the 100µm-filled and 150µm-filled vulcanizates

were relatively close when compared.

The values of flex fatigue decreases with increasing carbonization temperature of the mixes for all the fillers below 600°C before it starts rising again presented in TABLE 4 shown in Figure 6. A decrease in flex fatigue has been explained in terms of adherence of the filler to the polymer phase leading to the stiffening of the polymer chain and hence resistance to stretch when strain is applied<sup>[13]</sup>.

The abrasion resistance of a solid body is defined as its ability to withstand the progressive re-

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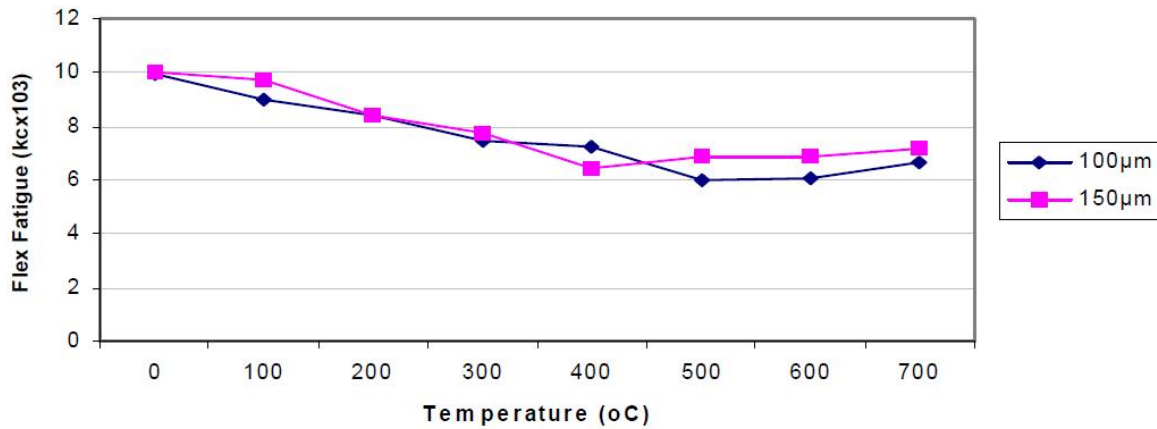


Figure 6 : Flex fatigue of 100µm-filled and 150µm-filled vulcanizates

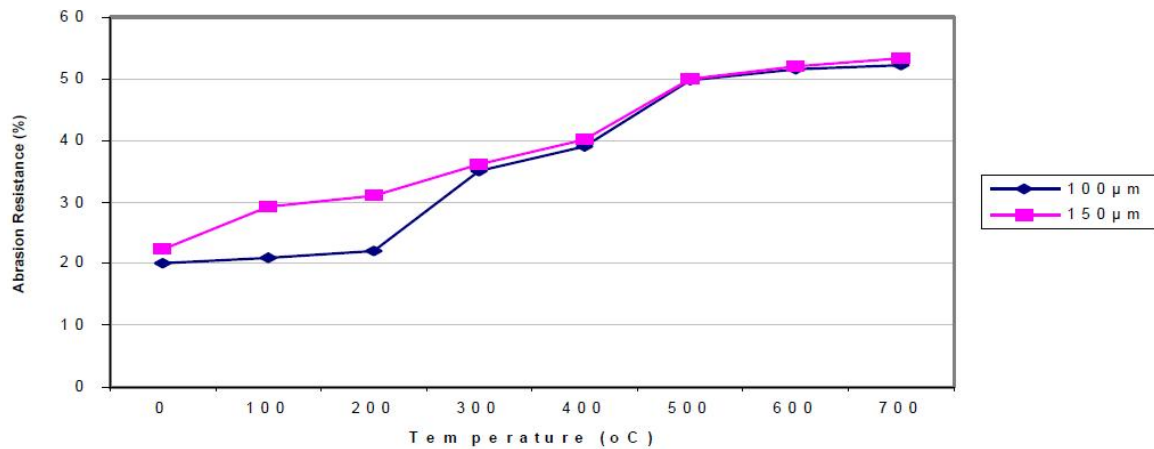


Figure 7 : Abrasion resistance of 100µm-filled and 150µm-filled vulcanizates

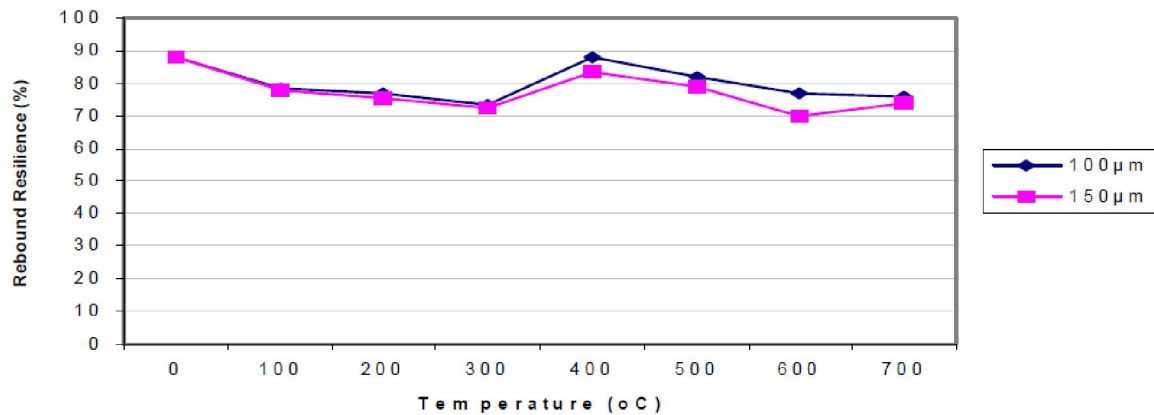


Figure 8 : Specific gravity of 100µm-filled and 150µm-filled vulcanizates

removal of the material from its surface as a result of the mechanical action of rubbing, scraping or erosive nature<sup>[14]</sup>. The trend of abrasion resistance with carbonization temperature of filler presented in TABLE 4 and Figure 7 show a regular pattern of increase with increasing the filler carbonization temperature for both the 100µm-filled and 150µm-filled vulcanizates. This indicates that filler carbonization temperature is a function of the measured parameter

attributed to the degree of dispersion of the fillers. However; the abrasion resistance of the vulcanizates decreases above 600°C, hence 100µm-filled has better abrasion resistance when compared with 150µm-filled vulcanizates.

The result of the specific gravity for the both 100µm-filled and 150µm-filled vulcanizates has showed in TABLE 4 increases and decreases with carbonization temperature. However; the 100µm-

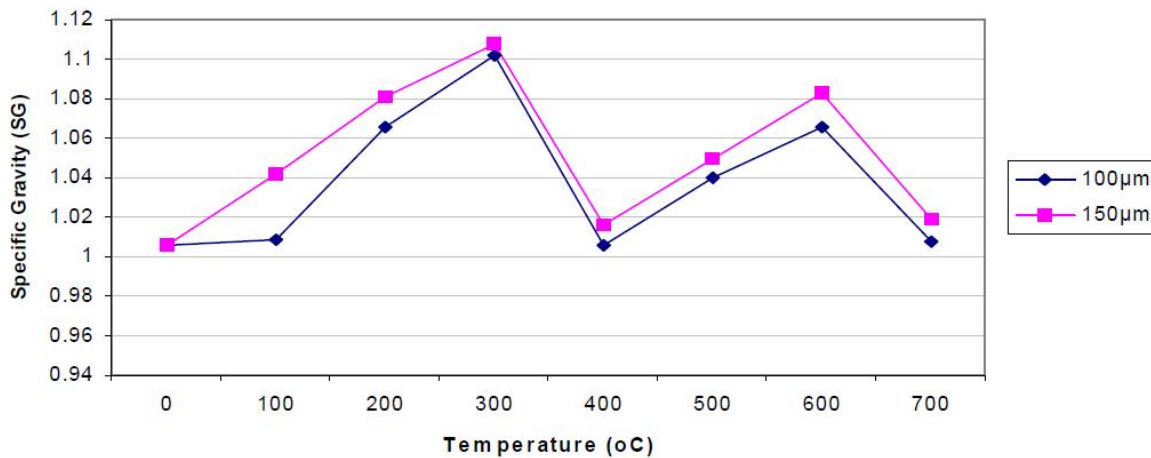


Figure 9 : Rebound resilience of 100µm-filled and 150µm-filled vulcanizates

filled vulcanizates show better stability than the 150µm-filled vulcanizates which may have been as a result of its compatibility with the rubber matrix.

The results of the vulcanizates presented in TABLE 4 and Figure 9 showed that 100µm-filled vulcanizates has excellent rebound resilience when stretched considerably than the 150µm-filled which has been explained in terms of filler adherence to the rubber matrix.

## CONCLUSION

The essence of this research work is to examine how the filler carbonization temperature of the palm kernel shells may influence its characteristics properties and hence the mechanical properties of the vulcanizates. The initial result showed that 100µm filler particle size is more reinforcing for natural rubber compound than 150µm. The results indicate that mechanical and swelling behaviour of the vulcanizates are greatly influenced by filler carbonization temperature and are therefore significant factors in determining the application in rubber compounding. The result also predicts the potential applications of CPKS as low cost filler in natural rubber products. The vulcanizates exhibit high quality characteristics at filler carbonization temperature (600°C). This indicate that for high quality vulcanizates using CPKS as reinforcing fillers, the particle size should be relative small and carbonization should be done at 600°C.

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