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### SORPTION PROPERTIES OF CARBONIZED/UNCARBONIZED CORNHUB POWDER FILLED NATURAL RUBBER/ACRYLONITRILE BUTADIENE RUBBER BIOCOMPOSITE

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#### ABSTRACT

The sorption characteristics of cornhub powder (carbonized/uncarbonized) filled natural rubber/acrylonitrile butadiene rubber (NR/NBR) using an aromatic solvent (toluene) have been studied. The effects of cornhub powder content, particle size, nature of solvent at temperature of 35°C were also investigated. The restriction on elastomer swelling exerted by the cornhub on the composite was investigated. Carbonized cornhub powder of 0.1µm particle size shows the lowest percentage swelling. The molecular percentage uptake of toluene for uncarbonized cornhub was higher than the carbonized cornhub. The effect of fibre loading swelling on the swelling behavior of the composite was also investigated in toluene. The percentage swelling index and swelling coefficient of the composite were found to decrease with increase in filler loadings and this shows an increased hindrance exerted by the fibre.

#### KEYWORDS

Sorption characteristics, Cornhub powder (carbonized/uncarbonized) and Natural rubber/acrylonitrile butadiene rubber (NR/NBR).

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#### INTRODUCTION

The demand for polymers in many applications has experienced steady growth over the years. In developed countries of Europe and America, the volumes of polymer used are more than that of metals or ceramics<sup>1</sup>. One of the most important phenomena in polymer science that has captured great research and industrial interest is the blending of polymers, since blending is a relatively simple way to create materials with significantly improved properties. Rubbers, being a major form of polymer

have not been left out. Different polymers may be blended to obtain a more cost effective material that combines the properties of both polymers that cannot be obtained from the individual polymers<sup>2</sup>.

Rubber in its raw state have virtually no engineering applications, it is necessary to appropriately compound them in order to obtain good processing characteristics, and the desired cure properties in the finished product<sup>3</sup>. Rubbers (Natural and Synthetic) have found numerous applications in our modern world. However these polymers are usually compounded with natural minerals or natural fibres to improve their mechanical properties as well as their sorption properties such as solubility, percentage swelling and swelling coefficients.

Natural rubber when vulcanized possesses unique properties such as high tensile strength, comparatively low elongation, hardness and abrasion resistance which found it's useful in the manufacture of various products. The main use of natural rubber is in automobiles. They can also be used in houses, footwears, battery boxes, balloons, toys and so many others.

The use of natural fibres as reinforcements or fillers in polymer composite systems has gained extra attention in recent years. Numerous studies have been carried out on the utilization of natural fillers such as sago, sisal, short silk fibre, oil palm, empty fruit bunch, rice husk ash, cornhub, jute fibre, rubber wood powders, hemp, kenaf and cellulosic fibres as reinforcement materials<sup>4</sup>. Compared with traditional reinforcement materials such as inorganic fibres, natural fibres such as cornhub have advantages such as low density, low cost, eco-friendly, abundance, renewable, less abrasive, high toughness and comparable specific strength, and biodegradable<sup>5</sup>.

The presence of solvents in polymers upon blending assumes significance since most polymers after swelling in the solvent show reduction in their properties. The effects of these solvents are believed to be due to localized plasticization that allows the development of crazes or cracks at reduced stress<sup>6</sup>. Therefore, polymers for commercial application should be chemically resistant and retain their mechanical integrity and dimensional stability on contacts with solvent<sup>7</sup>.

Many literature sources have revealed excellent reports on the sorption processes as well as mechanical properties of elastomer/thermoplastic blends. Polymers swell if they interact with the solvents, and the degree of this interaction is determined by the degree of crosslinks. The degree of swelling can be measured or related to the thermodynamic properties of the system<sup>8</sup>. Considerable interest has been focused on the absorption and diffusion of organic solvent because their ability to permeate at different rate enhances the separation of component of their liquid mixture through polymeric membrane<sup>9</sup>.

The diffusion characteristics of toluene, an aromatic solvent into natural rubber/linear low-density polyethylene blends were investigated by Obasi *et al.* (2009)<sup>10</sup>. The transport of toluene through most of the blend compositions was anomalous, although at 55°C, the transport of toluene through the 60/40 NR/LLDPE blend was Fickian, and at 35°C, pseudo-Fickian. The diffusion coefficient and permeability to toluene in 50/50 and 60/40 NR/LLDPE blends were found to increase with increased sorption temperature.

Similarly, Boonstra and Dannenberg (1958) presented the equilibrium swelling data of filled natural rubber and a number of synthetic rubbers in a variety of solvents. They observed that fillers such as carbon black showed a reduction in swelling of the membranes, and which was commensurate with the volume of the filler loading. However, the effects observed were not specific on a particular solvent or elastomer. The non-carbon black filler showed a reduction in rubber swelling, which was not dependent on the filler content.

Akporhonor *et al.*, (2007)<sup>11</sup> Investigated the equilibrium sorption properties of palm kernel husk and N330 filled natural rubber vulcanizates as a function of filler volume fraction. The result obtained showed that there was a decrease in sorption with increasing filler loading which he accounted on the basis of the fact that each filler particles behaves as an obstacle to the diffusing molecules. As concentration of filler increases in the rubber matrix, more and more obstacles are created

to the diffusing molecules which ultimately reduce the amount of penetrant solvent.

The effect of groundnut shell filler carbonizing temperature on the mechanical properties of natural rubber composite was studied by Ayo *et al.*, (2011)<sup>12</sup>. He found that the tensile strength, modulus, hardness and abrasion resistance increased with filler loadings while other properties such as compression set, flexural fatigue and elongation decreased with filler loadings. The percentage swelling in benzene, toluene and xylene were found to decrease with increased carbonization.

A study on the effect of carbon black powder on the physical properties of natural rubber/styrene butadiene rubber (NR/SBR) blends were carried out by Oleiwi *et al.*, (2011)<sup>13</sup>. In his study, 20 different rubber compounds were prepared by using SMR - 20 type of natural rubber and SBR - 1502 type of Styrene - butadiene rubber in different levels and each recipe reinforced with carbon black at different ratio. The physical properties such as thermal conductivity, thermal diffusivity, swelling and specific gravity were studied. The results showed that thermal diffusivity and thermal conductivity increased with increased carbon black and these properties decreased with increased NR/SBR blend. The swelling property measured by change in mass method; by immersing the samples in oil and water. Result showed that the swelling in water was more than in oil, change in mass of the NR/SBR blends increased with increase in immersing time and decreased with increased carbon black, and with increase in NR percentage to SBR. The specific gravity also increased with increased carbon black loading for all recipes.

Egwaikhide *et al.*, (2007)<sup>14</sup> Investigated the effect on the rheological, physic - mechanical and swelling properties of natural rubber vulcanizate using coconut fibre as fillers. The physic - mechanical properties as well as the equilibrium swelling characteristics of the vulcanizate in organic solvent were measured as a function of filler loadings and compared with the values obtained using commercial grade carbon black (N330). For coconut fibre filled vulcanizates, optimum tensile strength of 7.35MPa at 60ph was reported. Hardness of filled vulcanizates

increased with filler loadings. Abrasion resistance decreased marginally with increased filler loadings. Flexural resistance and percentage compression set decreased with increasing filler loading.

The equilibrium sorption in organic solvents of natural rubber vulcanizate filled coconut fibre and carbon black decreased with increased filler loadings. However, the resistance to swelling of natural rubber compound was dependent on the amount of filler loading: The higher the filler content, the lower the equilibrium sorption value obtained. Sorption characteristic of dynamically vulcanized polypropylene/epoxidized natural rubber blends filled with carbonized dikanut shell was studied by Onyeagoro and Enyiebulam (2012)<sup>15</sup>. It was observed that the resistance to toluene sorption increased with increased dosage of crosslinking agent due to increase in cross - link density as a result of increased network formation. Resistance to toluene sorption with increase in filler carbonization temperature was also observed. This was attributed to increase in surface area of the filler due to elimination of volatile matter that were deleterious to the blend - filler interfacial interaction.

Yakubu *et al.*, (2010)<sup>16</sup> Studied the physico - mechanical effects of surface-modified sorgum stalk powder on reinforced natural rubber, and found that fillers reduces the water absorption resistance which is in agreement with Ragumathen *et al.*, (2011)<sup>17</sup>. Ahmed *et al.* (2004)<sup>18</sup> Investigated the swelling properties of filled natural rubber/linear low-density polyethylene blends in toluene for 24 hour. He observed that the swelling index decreased with increased filler loading due to increased cross - link density of the blend.

In this work, blends of natural rubber/acrylonitrile butadiene rubber have been prepared using carbonized and uncarbonized corn hub powder filler of particle size 0.1 and 0.15 $\mu$ m. The sorption properties of the composites produced have also been studied. Also the effects of carbonization and filler loadings on the sorption properties of the composite have been studied and are detailed below.

## **EXPERIMENTALS**

### **Materials Used**

The natural rubber used in this study conforming to the Nigeria Standard Rubber Grade 10 (NSR 10) of specific density of 0.92 was obtained from Rubber research Institute, Iyanomo, Benin City, Edo state. The rubber vulcanizates were in form of rectangular sheet weighing 10kg with approximate thickness of 5mm. The acrylonitrile butadiene rubber (NBR) used in this study was obtained from Benin City, Edo State. The cornhub was obtained from Auchi, Edo state and sieved to 0.1 $\mu$ m and 0.15 $\mu$ m particle sizes respectively. The following weights (g) of cornhub powder were used in compounding the natural rubber/NBR blend 0, 10, 15, 20, 25, 30. The solvent used in this study was special purpose grade toluene of density 0.867g/ml and solubility parameter 8.91cal/ml.

Other rubber grade compounding ingredients used were stearic acid, zinc oxide, sulphur, tetramethyl thiarman disulphide (TMTD), trimethyl quinine (TMQ), processing oil (dioctylphthalates (DOP)), and carbon black and were purchase from a chemical store in Benin City.

### **Preparations of Cornhub Fillers and Carbonization**

The cornhub were washed in water and dried in air to remove sand particles and moisture respectively. After drying, half of the cornhub were milled to fine powder and sieved through a mesh size of 0.10 $\mu$ m and 0.15 $\mu$ m respectively. The fine particle that passes through the sieved were collected and used for compounding. The remaining half were weighed and carbonized in a Muffle furnace METTm - 525 at a temperature of 600°C. The carbonized cornhub were then milled to fine powder and sieved through a mesh size of 0.10 $\mu$ m and 0.15 $\mu$ m. The fine particle that passes through the mesh were collected and used for compounding.

### **Preparation of NR/NBR composite**

Three different sets of NR/NBR composites were prepared. NR/NBR with carbonized cornhub (CCH), NR/NBR with uncarbonized cornhub (UCCH), NR/NBR with carbon black composites were prepared at different filler loadings 0, 10, 15, 20, 25

and 30. The compositions of the NR/NBR composites are shown in the table below.

### **Characterization of the Filler**

The cornhub fillers used in this research were characterized for pH, bulk density, moisture content, iodine adsorption number and loss on ignition (LOI). The results obtained were comparable with that of carbon black (CB).

### **Characteristic of the solvent (toluene)**

See Table No.3.

### **Mixing Procedure (ASTMD 3184 - 80)**

The rubber mix were prepared on a laboratory size two roll mill according to the mixing formulation shown in Table No.1, it was maintain at 80°C to avoid cross linking during mixing after which the rubber composites were stretched out. The curing test pieces were done in a compression moulding machine. The machine was pre-set at a temperature of 140°C and a pressure of 10MPa, the curing time was 15 minutes.

### **Procedures for Sorption Test**

Blends of uniform sizes were cut and weighed on an analytical balance having an accuracy of 0.001g. The cut samples were put into sample bottles with covers. 20ml of toluene was poured into each of the sample bottles. The bottles were placed in a thermostatically controlled water bath at room temperature, 35°C for 24 hour. At the expiration of the specified time, the blends were removed from the sample bottles, wiped free of adhering toluene, and weighed using the analytical balance<sup>19</sup>. Each weighing was completed in less than 40 seconds, so as to keep the error due to solvent evaporation from the sample surface at a minimum.

## **RESULTS AND DISCUSSION**

### **Characterization of Carbonized and Uncarbonized Cornhub**

The pH changes from acidity to alkaline as the cornhub was carbonized due to residual material loss on combustion, the metallic content activity increases leading to alkalinity. This result was in agreement with Ayo et al (2011)<sup>12</sup> who studied the effect of carbonization temperature on the mechanical properties of groundnut shell filled natural rubber composite. The moisture content

decreased from 1.20% for uncarbonized cornhub to 0.08% for the carbonized filler. Since water boils at 100°C, it was suspected that water will vaporized and escape from the material leading to a low percentage of water in the filler.

The moisture content of filler is often used to predict the degree of defect arising from shrinkage during curing, particularly for products processed at elevated temperatures. This parameter indicates the macro structure of a fibre and reflects the reaction and adsorption abilities (Drivers, 1979)<sup>20</sup>. The high surface activity results from higher modulus at higher strain, higher abrasion resistance and lower hysteresis<sup>21</sup>. From the above table a difference in bulk density between the uncarbonized and the carbonized cornhub samples were seen. The bulk density for the carbonized cornhub was lower than the uncarbonized cornhub, this was as a result of the difference in particle size and structure of the fibre. The lower the bulk density, the better the interaction between the matrix and the filler, this will thus enhance the composite tensile strength and modulus.

#### **Sorption Properties of the Composite**

Table No.6 shows the sorption data presentation template for both blends (carbonized and uncarbonized biocomposite) carried out at a temperature of 35°C for 24 hours for the two particle sizes (0.1µm and 0.15µm) at different filler concentration (0, 10, 15, 20, 25, and 30). The sorption data of toluene into natural rubber/acrylonitrile butadiene rubber (NR/NBR) blend filled cornhub powder obtained at different filler loadings and particle sizes were determined and expressed as the percentage swelling of toluene per gram of NR/NBR blends.

#### **Percentage Swelling Index**

The percentage swelling index was calculated using the equation (Ayo et al, 2011)<sup>12</sup>

$$\text{Swelling Index \%} = (W_2 - W_1)/W_1 \times 100$$

Where  $W_1$  and  $W_2$  are the initial and final weights of the sample.

The percentage swelling index decreased with an increased filler concentration. The percentage swelling index was higher in uncarbonized cornhub (UCH) than the carbonized cornhub (CCH) and carbon black. The decrease in sorption with

increased filler concentration was attributed to filler particles which behave as an obstacle to the diffusing molecules. As the concentration of filler increases in the rubber matrix, more and more obstacles were created to diffusing molecules which ultimately reduced the amount of penetrant solvent. For the two fillers investigated, equilibrium sorption decreased with increased filler loading, suggesting a swelling restriction of the rubber matrix due to the presence of the filler. The swelling restriction caused by carbon black was smaller than that of the carbonized cornhub and uncarbonized cornhub; this was attributable to the better cross links formed by carbon black with the elastomer when compare to cornhub. The results were further explained in the figure below:-

#### **Swelling Coefficient**

The swelling behaviour of the composites can also be analyzed from the swelling coefficient values. It is an index of the ability with which the sample swells and this was determined using the equation

$$\text{Swelling coefficient, } \alpha = \{A_s/m\} \times (1/d)$$

Where,

$A_s$  is the weight of the solvent sorbed at equilibrium swelling,  $m$ , the mass of the sample before swelling and  $d$  the density of the toluene used (0.87 g/ml). The calculated values of the swelling coefficient ( $\alpha$ ) are presented in Table No.6.

The data in Table No.6 shows an increased filler loading with decreased swelling coefficient, While unvulcanized rubber dissolves in a good solvent, vulcanized rubbers can only be swollen in solvent to an extent determined by cross link density and the nature of the solvent (Akporhonor *et al*, 2007)<sup>11</sup>. The data was represented in Figure No.2. The decreased swelling coefficient values of the filled NB/NBR blends with increased filler loading indicates its reinforcing ability. The factors which can influence the equilibrium sorption in organic solvent of gum and filled vulcanizates, are nature of solvent and filler, level of cross-link and filler dispersion in the polymer matrix.

#### **Molecular % uptake of the Solvent**

The molecular % uptake of the solvent,  $Q_t$ , for the composite samples was determined using the equation (Mathew, 2002)<sup>22</sup>.

$$Q_t = \frac{\text{Mass of toluene absorbed}}{\text{Molecular weight of toluene/Initial mass of blend}} \times 100$$

The molar percentage uptake ( $Q_t$ ) of NR/NBR blends filled cornhub powder obtained at any particular filler loading was plotted against the filler loading of the composite for both the carbonized and uncarbonized blends. The molar percentage uptake ( $Q_t$ ) decreased with increased filler loadings.

The diffusion mechanisms in rubbers are essentially connected with the ability of the polymers to provide pathways for the solvent to progress in the form of randomly generated voids. As the void decreases with fibre addition, the solvent uptake also decreases. Figure No.3 shows the swelling behavior of the different composites in toluene. The composite showed the maximum swelling value indicating that there were more voids. It was observed that as filler loading increased, equilibrium solvent uptake decreased. This was obviously due to the increased hindrance exerted by the fibres at higher loading which was in accordance with Lovely *et al.*, (2005). Composites with low particle sized filler had more resistance than that with higher particle sized filler; the suspected trend was because the low particle sized filler had higher surface area and the interaction between the rubber matrix and fibre was higher, leading to higher crosslink density.

#### Crosslinking Density

The cross-link density of polymer i.e. the average molecular weight between the cross-links was determined from the swelling data.

$$\text{crosslinking density} = \frac{1}{Q}$$

Where  $Q = \frac{\text{swelling percentage}}{\text{molecular weight of toluene}}$

A comparison of crosslink density can be measured from the reciprocal swelling values  $1/Q$  (apparent crosslink density or crosslink value). The cross

linking density values were given in Table No.8. Increased cross linking density with increased in filler concentration and decreased filler particle sizes were observed. In this study, the cross linking density values for NR/NBR blends of both carbonized and uncarbonized cornhub (CCH and UCCH) were found to increase with increased filler loading but the CCH showed a significant increase. The order of cross linking density values is  $CB > CCH > UCCH$ .

Carbon black shows the highest crosslink value followed by CCH (0.1 $\mu$ m), CCH (0.15 $\mu$ m), UCH (0.1 $\mu$ m), UCH (0.15 $\mu$ m). This was as a result of average molecular weight of the polymers between crosslinks which was observed to decrease in their values. This may be attributed to increasing amount of filler in the rubber matrix. The molecular movement of the rubber reduces which made it more difficult for toluene to penetrate through the rubber matrix. The resistance to toluene sorption increased with increased dosage of crosslinking agent due to increased cross - link density as a result of increased network formation. Resistance to toluene sorption with carbonized cornhub filled NR/NBR vulcanizate was also observed. This was attributed to increased surface area of the filler due to elimination of volatile matter that were deleterious to the blend - filler interfacial interaction which was in agreement with Onyeagoro and Enyiebulam (2012)<sup>15</sup>. The cross-link density of polymer i.e. average molecular weight between the cross-links was determined from swelling data. A clear linear increase of the apparent cross-link density with the filler contents in the composite was observed indicating a strong interaction between the rubber matrix and the fillers which leads to strong physical cross-links.

**Table No.1: Compounding recipe for filler reinforced NR/NBR composites**

| S.No | Ingredients (phr)              | Formulation        |     |     |     |     |
|------|--------------------------------|--------------------|-----|-----|-----|-----|
|      |                                | A                  | B   | C   | D   | E   |
| 1    | Natural Rubber                 | 60                 | 60  | 60  | 60  | 60  |
| 2    | Acrylonitrile butadiene rubber | 40                 | 40  | 40  | 40  | 40  |
| 3    | Stearic acid                   | 2                  | 2   | 2   | 2   | 2   |
| 4    | Zinc oxide                     | 5                  | 5   | 5   | 5   | 5   |
| 5    | TMTD                           | 1                  | 1   | 1   | 1   | 1   |
| 6    | TMQ                            | 0.5                | 0.5 | 0.5 | 0.5 | 0.5 |
| 7    | Sulphur                        | 3                  | 3   | 3   | 3   | 3   |
| 8    | Processing oil                 | 5                  | 5   | 5   | 5   | 5   |
| 9    | Filler                         | (0,10,15,20,25,30) |     |     |     |     |

**Table No.2: Characterization of Rubbers**

| S.No | Properties       | Natural rubber | NBR rubber |
|------|------------------|----------------|------------|
| 1    | Specific density | 0.92           | 0.95       |
| 2    | Temp. range °C   | -51 to 104     | -30 to 107 |
| 3    | Elongation       | >300%          | >500%      |

**Table No.3: Characteristic of the toluene (solvent)**

| S.No | Parameters                    | Characteristic of toluene |
|------|-------------------------------|---------------------------|
| 1    | Grade                         | Special purpose grade     |
| 2    | Density (g/ml)                | 0.867                     |
| 3    | Solubility parameter (cal/ml) | 8.91                      |
| 4    | Molar mass volume (g/mol)     | 92.14                     |

**Table No.4: Characterization of cornhub filler and carbon black**

| S.No | Parameter                         | Cch  | Ucch | Carbon black |
|------|-----------------------------------|------|------|--------------|
| 1    | pH of slurry at 28°C              | 7.86 | 5.20 | 6.00         |
| 2    | Moisture content (%)              | 0.08 | 1.20 | 0.42         |
| 3    | Loss on Ignition (%)              | 79.3 | 82.5 | 91.3         |
| 4    | Bulk density (g/cm <sup>3</sup> ) | 0.61 | 0.82 | 0.40         |

**Table No.5: Percentage Swelling Index of the Composite**

| S.No | Filler Loading (wt. %) | CCH (0.1µm)    |       | UCCH (0.1µm)   |       | CCH (0.15µm)   |       | UCCH (0.15µm)  |       | Carbon black   |       |
|------|------------------------|----------------|-------|----------------|-------|----------------|-------|----------------|-------|----------------|-------|
|      |                        | W <sub>1</sub> | PS    | W <sub>1</sub> | PS    | W <sub>1</sub> | PS    | W <sub>1</sub> | PS    | W <sub>1</sub> | PS    |
| 1    | 0                      | 0.910          | 400.4 | 0.910          | 400.4 | 0.910          | 400.4 | 0.910          | 400.4 | 0.910          | 400.4 |
| 2    | 10                     | 0.920          | 290.8 | 0.912          | 358.4 | 0.916          | 320.0 | 0.920          | 365.8 | 0.917          | 269.3 |
| 3    | 15                     | 0.917          | 284.6 | 0.918          | 344.2 | 0.913          | 312.8 | 0.913          | 360.2 | 0.908          | 260.0 |
| 4    | 20                     | 0.915          | 270.8 | 0.915          | 332.6 | 0.920          | 302.6 | 0.918          | 340.0 | 0.914          | 248.1 |
| 5    | 25                     | 0.910          | 266.8 | 0.913          | 328.4 | 0.918          | 286.4 | 0.912          | 336.2 | 0.925          | 240.0 |
| 6    | 30                     | 0.912          | 260.4 | 0.916          | 320.5 | 0.914          | 279.3 | 0.914          | 330.0 | 0.916          | 236.4 |

\*ps= percentage swelling index

**Table No.6: Swelling Coefficient**

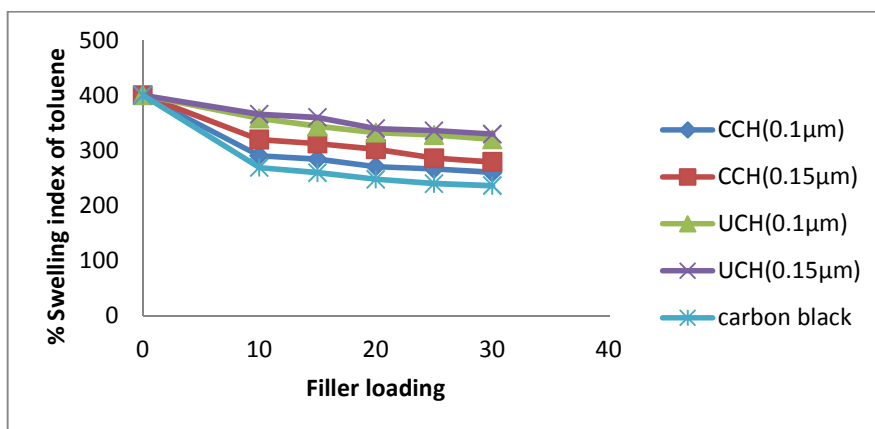
| S.No | Filler loading | CCH (0.1µm) |       | UCCH (0.1µm) |       | CCH (0.15µm) |       | UCCH (0.15µm) |       | Carbon black |       |
|------|----------------|-------------|-------|--------------|-------|--------------|-------|---------------|-------|--------------|-------|
|      |                | m           | α     | M            | α     | m            | α     | M             | α     | M            | α     |
| 1    | 0              | 0.910       | 4.601 | 0.910        | 4.601 | 0.910        | 4.601 | 0.910         | 4.601 | 0.910        | 4.601 |
| 2    | 10             | 0.920       | 3.341 | 0.912        | 4.118 | 0.916        | 3.677 | 0.920         | 4.203 | 0.917        | 3.094 |
| 3    | 15             | 0.917       | 3.270 | 0.918        | 3.955 | 0.913        | 3.594 | 0.913         | 4.139 | 0.908        | 2.987 |
| 4    | 20             | 0.915       | 3.112 | 0.915        | 3.822 | 0.920        | 3.477 | 0.918         | 3.907 | 0.914        | 2.851 |
| 5    | 25             | 0.910       | 3.066 | 0.913        | 3.773 | 0.918        | 3.291 | 0.912         | 3.863 | 0.925        | 2.758 |
| 6    | 30             | 0.912       | 2.992 | 0.916        | 3.683 | 0.914        | 3.209 | 0.914         | 3.792 | 0.916        | 2.716 |

**Table No.7: Molecular % Uptake of Toluene**

| S.No | Filler loading | CCH (0.1µm)    |                                | UCCH (0.1µm)   |                                | CCH (0.15µm)   |                                | UCCH (0.15µm)  |                                | Carbon black   |                                |
|------|----------------|----------------|--------------------------------|----------------|--------------------------------|----------------|--------------------------------|----------------|--------------------------------|----------------|--------------------------------|
|      |                | Q <sub>t</sub> | W <sub>2</sub> -W <sub>1</sub> | Q <sub>t</sub> | W <sub>2</sub> -W <sub>1</sub> | Q <sub>t</sub> | W <sub>2</sub> -W <sub>1</sub> | Q <sub>t</sub> | W <sub>2</sub> -W <sub>1</sub> | Q <sub>t</sub> | W <sub>2</sub> -W <sub>1</sub> |
| 1    | 0              | 4.346          | 400.4                          | 4.346          | 400.4                          | 4.346          | 400.4                          | 4.346          | 400.4                          | 4.346          | 400.4                          |
| 2    | 10             | 3.156          | 290.8                          | 3.889          | 358.4                          | 3.473          | 320.0                          | 3.970          | 365.8                          | 2.923          | 269.3                          |
| 3    | 15             | 3.088          | 284.6                          | 3.736          | 344.2                          | 3.395          | 312.8                          | 3.909          | 360.2                          | 2.822          | 260.0                          |
| 4    | 20             | 2.939          | 270.8                          | 3.609          | 332.6                          | 3.284          | 302.6                          | 3.690          | 340.0                          | 2.693          | 248.1                          |
| 5    | 25             | 2.896          | 266.8                          | 3.564          | 328.4                          | 3.108          | 286.4                          | 3.649          | 336.2                          | 2.605          | 240.0                          |
| 6    | 30             | 2.826          | 260.4                          | 3.478          | 320.5                          | 3.031          | 279.3                          | 3.581          | 330.0                          | 2.566          | 236.4                          |

**Table No.8: Crosslinking Density**

| S.No | Filler loading | CCH (0.1µm) |       | UCCH (0.1µm) |       | CCH (0.15µm) |       | UCCH (0.15µm) |       | Carbon black |       |
|------|----------------|-------------|-------|--------------|-------|--------------|-------|---------------|-------|--------------|-------|
|      |                | Q           | 1/Q   | Q            | 1/Q   | Q            | 1/Q   | Q             | 1/Q   | Q            | 1/Q   |
| 1    | 0              | 4.346       | 0.230 | 4.346        | 0.230 | 4.346        | 0.230 | 0.910         | 0.230 | 0.910        | 0.230 |
| 2    | 10             | 3.156       | 0.317 | 3.889        | 0.257 | 3.473        | 0.288 | 0.920         | 0.252 | 0.917        | 0.342 |
| 3    | 15             | 3.088       | 0.323 | 3.736        | 0.268 | 3.395        | 0.293 | 0.913         | 0.256 | 0.908        | 0.354 |
| 4    | 20             | 2.939       | 0.340 | 3.609        | 0.277 | 3.284        | 0.305 | 0.918         | 0.271 | 0.914        | 0.371 |
| 5    | 25             | 2.896       | 0.344 | 3.564        | 0.281 | 3.108        | 0.322 | 0.912         | 0.275 | 0.925        | 0.384 |
| 6    | 30             | 2.826       | 0.354 | 3.478        | 0.288 | 3.031        | 0.330 | 0.914         | 0.279 | 0.916        | 0.390 |



**Figure No.1: Plot of percentage swelling index of toluene versus filler content for NR/NBR at different filler particle sizes**



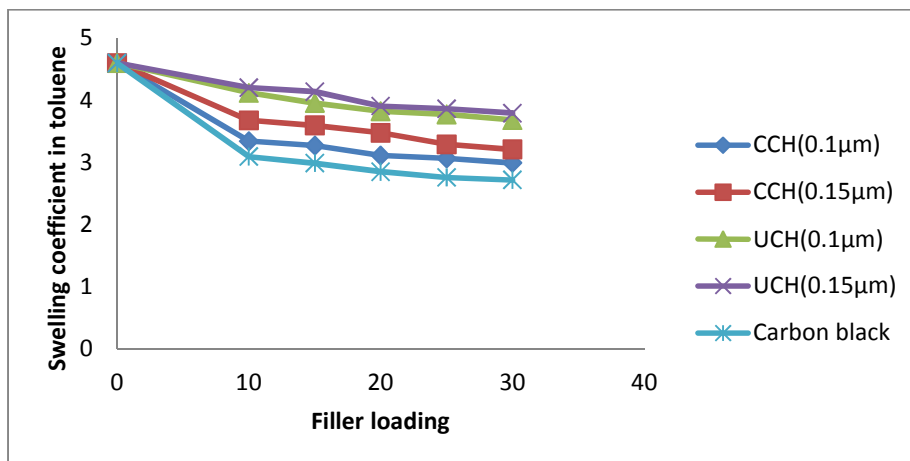


Figure No.2: Plot of swelling coefficient of toluene versus filler content for NR/NBR at different filler particle sizes

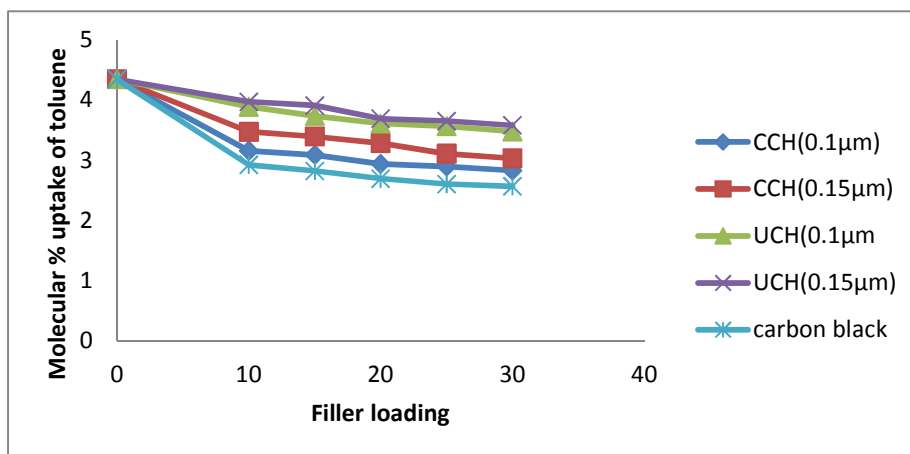


Figure No.3: Plot of molecular percentage uptake of toluene versus filler content for NR/NBR at different filler particle sizes

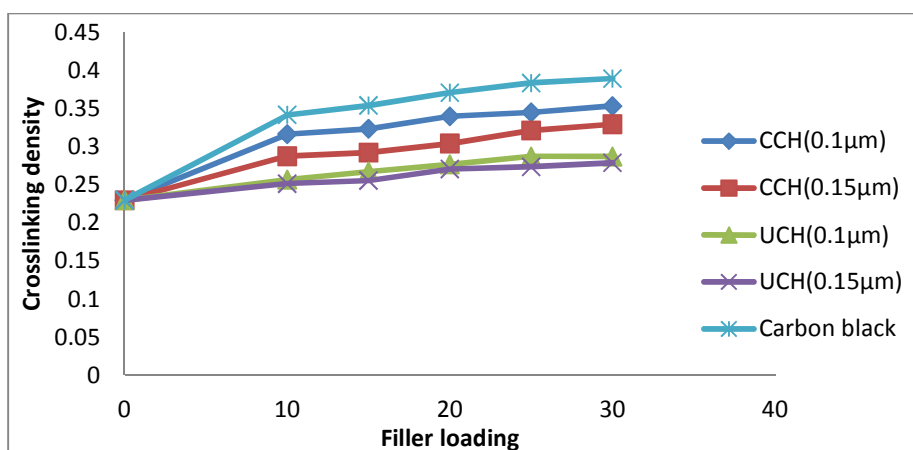


Figure No.4: Plot of crosslinking density of toluene versus filler content for NR/NBR at different filler particle sizes

## CONCLUSION

The sorption behaviour of toluene through cornhub powder filled Natural rubber/acrylonitrile butadiene rubber (NR/NBR) blend at different filler loadings and different particle sizes had been studied. The percentage swelling was found to decrease with increased filler loading for the two particle sizes studied, with UCH (0.15 $\mu$ m) showing the highest swelling restriction ratio while carbon black shows the lowest swelling restriction ratio. The molecular % uptake was found to decrease with increased filler loading. Maximum uptake was observed with UCH followed by CCH and then Carbon black. Carbon black was observed to show the highest crosslink density followed by CCH and UCH respectively, this was because cross link density increased with increased filler loading. The solubility of the NR/NBR blend was observed to depend on the nature of the filler and the particle size. CCH of 0.1 $\mu$ m was observed to show better properties when compare to CCH (0.15 $\mu$ m) and UCH of various particle size but carbon black was slightly better.

Agricultural residues as by-products and co-products of agriculture and processing of agricultural product represent a large feedstock of underutilized resources which can be used directly or converted by fairly simple chemical processes into higher value added material. Cornhub fibre is an agricultural by-product of corn. The cost of cornhub fibre is relatively cheap compared to carbon black filled Natural rubber/NBR matrix in the cellulosic-based composite. Generally the level of property improvement observed in the swelling restriction shown by NR/NBR/cornhub biocomposite was encouraging. Cornhub fibre is hoped to definitely develop its niche in the filler market in future.

Furthermore, cornhub used as filler in the present study is a biodegradable agricultural by-product and thus, confers biodegradability to the NR/NBR blends. This will help to reduce the impact of non-biodegradable materials on our solid waste stream, which is already a global environmental problem compounded by the ever decreasing availability of landfill space as well as increasing cost of municipal solid waste disposal. This in addition to the production cost reduction is advantageous over the

use of convectional, but costly petroleum based reinforcing fillers such as carbon black.

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## CONFLICT OF INTEREST

We declare that we have no conflict of interest.

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