# **TENSILE PROPERTIES OF ALKALISED JUTE FIBRES**

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The structure of jute fibre has been modified using caustic soda up to a limit in order to improve its performance. The SEM micrographs of untreated jute fibres show a smooth surface, while alkalised jute fibres show rough and void regions between individual fibre cells. The study showed that the tensile strength and Young's modulus of jute fibre bundles depends on the physical characteristics of its internal structure such as the cellulose content, changes in the crystalline region content expressed in terms of crystallinity index, and micro-fibril angle. Results also showed tensile properties optimised at 0.24% NaOH (w/w). Overall, alkalised fibres exhibit brittle fracture. The study demonstrated the dependence of tensile properties on the changes in fibre structure following alkalisation.

*Keywords:* Fibres; Jute fibre; Mechanical properties; Natural fibre; Swelling; Structure-property relations

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# INTRODUCTION

The strength of plant fibres is attributed to the rigidity and high molecular weight of cellulose chains, intermolecular and intramolecular hydrogen bonding, and the fibrillar and the crystalline structure of the fibres (Nguyen et al. 1981). The strength and stiffness of fibres has also been shown to be dependent on the crystallinity index and micro-fibril angle. McLaughlin and Tait (1980) have showed that strain is more dependent on the micro-fibril angle and that it increases with an increase in the micro-fibril angle. Fibres with higher cellulose content have also been found to be stronger than those with a low cellulose content as long as their micro-fibril angle is small (Morton and Hearle 1975). For instance, cotton with a micro-fibril angle of 20 to 30° and cellulose content of over 90% exhibits lower mechanical properties than fibres which have micro-fibril angles of less than 7° and cellulose content of over 80%, such as hemp and flax fibres (Morton and Hearle 1975; Mwaikambo 2002). The above-mentioned characteristics make these fibres attractive materials for end uses such as reinforcements for polymeric materials.

The mechanical properties reflect the orientation of the micro-fibrils, which are inclined at an angle to the cell axis. It is worth mentioning that fibrous materials are commonly subjected to the following deformations: tension, compression, bending, torsion, shear, abrasion, wear, and flexing (Morton and Hearle 1975) either while still in the plant source or in use following extraction from the plant.

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#### Modelling the Stiffness of the Cellulose Cell Wall

There is a strong correlation between the micro-fibril angle and the Young's modulus of the fibres. Models indicate that fibre stiffness is influenced by the spiral angle of the crystalline fibrils as well as the concentration of non-crystalline materials (Gassan and Bledzki 1999). These structural parameters vary between the different types of natural fibre, accounting for some of the variations in reported fibre properties. The effect on the mechanical properties of increased micro-fibril angle plays an important role when determining the mechanical properties of fibre-reinforced composites. It is necessary to measure the alignment of micro-fibrils applied to plant fibres to the direction of the force, especially in determining tensile properties, bearing in mind that plant fibres exhibit significant mechanical anisotropy. A theoretical analysis of the way a fibre behaves when stretched may in practice represent the behaviour of fibre-reinforced composites when determining their mechanical properties. In this case a uniform strain theory has been used to obtain an estimate of the stiffness of the entire arrangement in both the fibre and the cell wall composite material. The theory uses an assembly of springs tied together so that they all receive the same displacement  $\mu$  under a tensile load F, as shown in Figure 1. The theory is based on the work of Bodig and Javne (1982).



Fig. 1. Arrangements of a uniform elastic deformation model in the plant fibre cell wall

The quantities  $\mu$  and  $\dot{F}$  are the mean displacement and load on the micro-fibrils respectively.  $K_1$  and  $K_2$  are the micro-fibril constants. Let F be the load and  $\mu$  the displacement under tension. The total force necessary to produce the displacement is the sum of the forces in the fibre,  $F_f$ , applied to the crystalline and amorphous regions (eq. 1),

$$F_{f} = F_{c} + F_{nc} \tag{1}$$

where  $F_c$  and  $F_{nc}$  represent the forces applied to the crystalline and non-crystalline materials respectively. The stiffness (eq. 2) of the array is given as:

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$$k = \frac{F}{\mu} = \sum K_i$$
<sup>(2)</sup>

The components in Figure 1 are at an angle  $\theta$  to the direction of the force applied. A displacement  $\mu$  is applied in the direction of the applied force  $\overline{F}$ . The forces, which appear in each spring element representation, are obtained from the product of the stiffness and the component of the displacement that is parallel to their orientation (eq. 3).

$$\overline{F_i} = K_i \mu \cos \theta_i \tag{3}$$

The total force in the longitudinal direction of the fibre necessary to cause this displacement is the sum of the longitudinal forces on each crystallite (eq. 4).

$$\overline{F} = \sum F_i \cos \theta_i \tag{4}$$

Substituting  $F_i$  from above gives equation 5.

$$\overline{F} = \overline{\mu} \sum k_i \cos^2 \theta_i \tag{5}$$

The overall stiffness of the system of the fibre in the fibre axis is given by equation 6.

$$\bar{\mathbf{k}} = \sum \mathbf{K}_{i} \cos^{2} \theta_{i} \tag{6}$$

If  $\overline{k}$ = overall stiffness of the fibre (E<sub>f</sub>) and K<sub>i</sub> is the stiffness of the micro-fibrils (E<sub>s</sub>), then equation (6) becomes (7).

$$E_{f} = E_{s} \cos^{2}\theta \tag{7}$$

Equation 7 gives the longitudinal Young's modulus of plant fibres measured in tension. Efforts are being made to determine the transverse Young's modulus of plant fibres, but this is not covered here. However, Cichocki and Thomason (2002) used an environmental scanning electron microscopy (ESEM) equipped with a load frame to measure the Young's modulus of a carefully sectioned array of wood cells in the transverse direction. Their measurement indicated that the transverse Young's modulus varied with the type of wood used and was less than the longitudinal Young's modulus Cichocki and Thomason (2002).

#### Modelling of the Cell wall as a Composite Material

Since both the crystalline and non-crystalline region will deform, the applied load is shared between these two components, just as is the case with the composites. The determination of stiffness properties of plant fibres can be predicted by using the rule of mixtures (ROM). For instance, equation 7 is used to estimate the stiffness or modulus of elasticity of the plant fibre cell wall along the fibre axis (Bodig and Jayne 1982). The effective modulus of the fibre ( $E_f$ ) is given in terms of the amounts of participating components present in the fibre. The ROM takes into consideration the masses of the crystallites and non-crystallites to determine the stiffness of the composites (eq. 8),

$$E_{f} = V_{c}E_{c}\cos^{2}\theta + V_{nc}E_{nc}$$
(8)

where,  $E_c$  and  $E_{nc}$  are the moduli of crystalline and non-crystalline regions and  $V_c$  and  $V_{nc}$  are the volume fractions of crystalline and non-crystalline regions. Kulkarni et al. (1983) reported elastic moduli of the crystalline and non-crystalline regions for vegetable fibres as 45 GPa and 3 GPa respectively. When banana fibre was tested at a micro-fibril angle of 12° and 11°, with a fibre diameter of 100 µm with volume fractions of 0.65 and 0.35 for crystalline and non-crystalline components respectively, the modulus values obtained using equation 8 compared well with the practical modulus values obtained by Kulkarni et al. (1983) when determining the mechanical properties of banana fibres *Musa sepientum*. Mukherjee et al. (1993) performed similar work on sisal fibre tested at different test gauge lengths and speeds and the results obtained were comparable to the theoretical predictions.

### **Fibre Modifications**

Plant fibres must exhibit a compatible surface morphology for the development of a coherent interface with matrix polymers, but because of the surface impurities present in plant materials the development of a good fibre-matrix interface is impaired. However, in order to make use of the good physical and structural characteristics of the fibres, they are treated with chemicals such as caustic soda to modify surface topography and the fine structure (Avela et al. 1999; Mwaikambo and Ansell 1999; Mwaikambo and Ansell 2002; Mwaikambo and Ansell 2003). The main requirement for reinforcement is to use stiff fibres and a well-bonded fibre-matrix interface, with the ultimate aim of reaching the stiffness of conventional synthetic reinforcement such as carbon and glass fibres. In plant fibres these requirements are partially attained by alkali treatment. Jute fibres possess surface impurities such as wax and natural oils and sometimes processing oils can also be deposited at the surface. Bleaching and/or scouring using solvents can remove these surface impurities. Several fibre modification processes in textile manufacturing require the removal of these surface impurities to improve lustre and dye In composite manufacture removal of surface waxy materials improves uptake. mechanical interlocking and reactivity with the resins, thus developing strong interfacial adhesion (Avela et al. 1999; Mwaikambo and Ansell 1999; Mwaikambo and Ansell 2002; Mwaikambo and Ansell 2003). Cellulose forms the main structural constituent of plant fibres and contributes immensely to the mechanical properties of plant fibres. Other components such as lignin and hemicelluloses play an important part in the characteristic properties of the fibres. Toughness, which is the tendency of plant fibres to absorb energy in impact, is decreased with a decreasing amount of lignin and/or hemicelluloses, while at the same time the strength and stiffness of the fibre is increased up to a limit.

The development of fibre-reinforced composites favours the use of stiff fibres for reinforcement of polymeric materials (Avela et al. 1999). Removing lignin and hemicelluloses, thus leaving stiffer cellulose, can produce stiff plant fibres. The removal of hemicelluloses leaves a less dense and less rigid interfibrillar region, allowing the fibrils to re-arrange along the fibre major axis Murkherjee et al. 1993). Stretching the fibre results in better load sharing by the fibrils, hence higher stress development in the fibre. On the other hand, softening of the interfibrillar matrix adversely affects the stress transfer between the fibril and thereby the overall stress development in the fibre under tensile deformation. Removing the lignin makes the middle lamella joining the ultimate cells become more plastic and homogeneous due to the gradual elimination of microvoids whereas the ultimate cells themselves are only slightly affected.

The rearrangement of the fibrils along the fibre axis and the resulting homogeneity of the ultimates leads to a packing order with increased crystallinity index for the plant fibres. Physical methods such as heating/drying can be applied to plant fibres to remove hemicelluloses, which are sensitive to high temperatures. The steam explosion technique is another method commonly used to produce clean fibres. Several chemical methods have been applied to modify plant fibres and these are discussed in the following sections together with physical methods.

In this work jute fibre bundles that have been alkalised have been tested in tension and the results compared with conventional synthetic fibre reinforcements to assess the potential of jute fibre as an alternative to synthetic fibres. Also, the theoretical stiffness of plant fibres has been predicted and results compared with experimental values.

# **EXPERIMENTAL METHODS**

### **Materials**

Jute fibre used in this work originated, from Bangladesh. No specifications were available regarding the physical characteristics of the supplied fibres such as staple length, density, diameter, and processing conditions. Sodium hydroxide pellets of 98% concentration and glacial acetic acid were supplied as general laboratory reagents.

# Fibre Preparation

Fibres were stored in a conditioning chamber containing a saturated sodium nitrite solution whereby 85 g of the solute was added to 100 cm<sup>3</sup> of water. For a room temperature of  $20 \pm 2^{\circ}$ C, the conditioning chamber with the solution in it generates a relative humidity inside the chamber of  $65 \pm 2\%$  relative humidity.

# Alkali Treatment

Jute fibre bundles were soaked in beakers containing 0.03%, 0.08%, 0.16, 0.24%, and 0.32% weight by weight (w/w) concentrations of caustic soda (NaOH) and placed in a water bath controlled at  $20 \pm 2^{\circ}$ C for 48 hours. About 50g of jute fire bundles were submerged in 200ml of caustic soda. The 16% NaOH was used to study the effect of extreme concentration of caustic soda on fibres. The fibres were then removed, washed with distilled water containing 1 % acetic acid, to neutralise excess sodium hydroxide,

and then thoroughly rinsed with distilled water. The fibres were then dried to remove free water and placed in a glass container in a conditioning chamber.

### **Determination of Crystallinity index**

Untreated and treated fibres were mixed with a very small amount of an adhesive material Tragacanth BP, soaked in a drop of distilled water, compressed into sheets, and dried. A wide-angle X-ray diffractometer (WAXRD) equipped with a scintillation counter and a linear amplifier was used. The diffraction intensities were recorded between 5 and 60° (20-angle range). The crystallinity index (I<sub>c</sub>) was determined by using equation 9 where  $I_{(002)}$  is the counter reading at peak intensity at a 20 angle close to 22° representing crystalline material and  $I_{(am)}$  is the counter reading at peak intensity at a 20 angle close to 18° representing amorphous material in cellulose fibres.

$$I_{c} = \frac{\left(I_{(002)} - I_{(am)}\right)}{I_{(002)}} \times 100$$
(9)

### **Determination of Fibre Diameter**

Twenty-five fibre specimens each 20 mm long were cut and prepared for SEM examination. The SEM micrographs prepared were then subjected to image analysis and their diameter determined along the width of the fibre bundles. Fifty readings were recorded for each fibre specimen and the mean, standard deviation and standard error were calculated.

### **Determination of the Cellulose Content of Fibres by Density Methods**

The cellulose content of plant fibres was calculated using a combination of the bulk, absolute, and cellulose densities using equation 10. A full description of the way densities were determined is provided in Mwaikambo and Ansell (2001) and will not be repeated in this paper.

$$Z = \left[2\left(\frac{\rho_{b}}{\rho_{a}} + \frac{\rho_{a}}{\rho_{cell}}\right) - \frac{\rho_{b}}{\rho_{cell}} - 2\right]100$$
(10)

In this equation  $\rho_a$ ,  $\rho_b$ ,  $\rho_{cell}$  and Z are the absolute, bulk (apparent), cellulose densities, and cellulose content respectively.

### **Tensile Test**

The Instron tensile tester, model 1122, was used. The instrument incorporates a highly sensitive electronic load weighing system, with load cells employing strain gauges to detect the load applied to the specimen under test. For single ultimate fibres the most sensitive load cell is that with a capacity of 0.5N. However, because what were being tested are bundles of fibres in the case of jute fibre a load cell with a sensitivity of 10N was employed. This was calibrated by hanging a 1-kg weight on the upper jaw. With the pen switched on, the pen moves to full-scale deflection, and to acquire the highest sensitivity it was multiplied by a factor of ten. It was then adjusted to 9.81 divisions on the chart to compensate for the conversion of the gramme weight to load in newtons. The

pen was then switched off and the calibration weight was removed. The gripping jaws were adjusted to the length of the card. The crosshead speed was adjusted to give a standard time to break of about 20 seconds. The chart speed was then adjusted to give a reasonably sized deflection on the chart paper.

Fibres were temporarily fixed on the card by adhesive tape across a circular hole of 19mm diameter cut in a card. Figure 2 shows a cross-sectional view of the mounting card of a fibre test piece. A 'blob' of epoxy resin was dropped on the edge on both sides of the centre of the hole along the length of the card. In order to let the epoxy resin set, the whole set-up was left in the conditioning chamber for 48 hours before testing.



Fig. 2. Mounting card of a fibre test piece

The card was placed between the Instron jaws and a pair of scissors was used to cut both sides of the card as shown in Fig. 2 to start testing. Thirty-two specimens were considered sufficient for each test. The mean ultimate tensile strength, mean breaking strain and extension per cent, the mean Young's modulus and the respective standard deviation were determined. However, for comparing different fibres, the value of specific strengths on the basis of area of cross-section, the stress at break is termed the ultimate tensile strength. Tests for the tensile strength of the 16% NaOH treated jute fibre bundles were not performed due to the crimpy nature of the fibres following caustic soda treatment.

### **RESULTS AND DISCUSSION**

### Surface Topography and Transverse Sections

Figures 3a and b show longitudinal and cross sectional views respectively of untreated jute fibre bundles. The cross sectional view (Fig. 3b) shows small (indicated by S-arrow) and large (indicated by L-arrow) lumens, each representing a single ultimate fibre. Following 0.24% NaOH treatment (Fig. 4a) individual fibre cells are seen. The separation of the ultimates indicates that the binding materials have been removed. The cross sectional view of the 0.24% NaOH treated jute fibre bundles (Fig. 4b) shows clear absence of the lumen, indicating complete swelling of the cell wall. The closure of the

lumen due to swelling following 0.24% NaOH treatment can be attributed to two possible scenarios, namely that jute fibre possesses low lignin-cellulose interfacial energy (a factor that has not been experimentally determined in this work) and that a high concentration of lignin is between the primary wall and  $S_1$  layer as illustrated in Fig. 6. Furthermore the inter-microfibrillar lignin in the secondary layers is evenly distributed (Preston 1963). The progressive swelling mechanism of the cell wall during alkalisation is discussed in a later section of this paper and hypothetically illustrated in scheme 1.



Figure 3. SEM micrograph of (a) longitudinal view of untreated jute fibre bundles, and (b) cross sectional view of untreated jute fibre bundles



**Figure 4.** SEM micrograph of (a) longitudinal view of 0.24% NaOH treated jute fibre bundles, and (b) cross sectional view of 0.24% NaOH treated jute fibre bundles

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**Figure 5.** SEM micrograph of (a) longitudinal view of 16% NaOH treated jute fibre bundles, and (b) cross sectional view of 16% NaOH treated jute fibre bundles

Figures 5a & b show longitudinal and cross sectional views of 16% NaOH jute fibres bundles respectively. The longitudinal view of the 16% NaOH treated jute fibre bundles (Fig. 5a) show a distinct separation of ultimate fibres (arrows without letters). Some of these ultimates exhibit a ruptured cell wall indicated by arrows (R-arrows). The fusing and rupture of the ultimates indicates the severe effect of the 16% NaOH concentration on the cell wall. The cross-sectional view shows no sign of the lumen. The cells are believed to undergo inward swelling, and in some of them the hexagonal shape has started to disappear. This implies that caustic soda has attacked and started to depolymerise the crystalline region in the S<sub>1</sub>, S<sub>2</sub>, and S<sub>3</sub> layers. The differences in swelling within the cell wall will have significant impact on the mechanical properties of jute fibres. The cross-sectional view (Fig. 5b) shows no sign of the lumen indicating a complete swelling of the cell wall. Figure 6 shows a hypothetical impression of the cross-sectional view of untreated jute fibre with the positioning of the lignin within the cell wall layers.





#### Effect of the Fibre Diameter on the Tensile Properties

Following alkalisation the dependence of tensile strength and Young's modulus on the diameter of jute fibre bundles is illustrated in Figure 7 and Figure 8. The diameters and tensile properties of untreated and alkali treated jute fibre bundles were determined as described in section 2.

The high strength and modulus (Figs. 7 and 8 respectively) of jute fibre bundles with small diameter is understandable since, in the limit, a single unbroken chain of cellulose molecules must be approaching the theoretical tensile strength and stiffness of bonds between atoms (Mwaikambo 1984). Furthemore, fibre bundles with small diameter have reduced porosity and weak links.



Figure 7. The dependence of strength of jute fibre bundles on the fibre bundle diameter



Figure 8. The dependence of modulus of jute fibre bundles on the fibre bundle diameter

It is observed that the separation of ultimate fibres results in optimised tensile strength and Young's modulus at around 0.24% NaOH treatment (Figs. 7 and 8, respectively). The increase in diameter and, tensile strength (Fig. 7) and Young's modulus (Fig. 8) between untreated and 0.03% NaOH treatment are due to the swelling of the primary wall with no consequential degradation of the cell wall (Mwaikambo and Ansell 2006 a and b). In fact, at this concentration of caustic soda, lignin present between the primary wall and the  $S_1$  layer inhibits inward swelling of the cell wall (Preston 1963). This level of caustic soda concentration (0.03% NaOH) has a bleaching effect and removes all the impurities on the surface of the fibre. It was observed that between 0 and 0.03 % NaOH treatment pectin will be degraded and together with lignin will be removed from the cell wall causing separation of the ultimate fibres, thus decreasing the diameter of the bundle (Mukherjee et al. 1983). The decrease in diameter led to increase in the tensile strength and Young's modulus (Fig. 7 and 8). It was further observed that between 0.08% and 0.16% NaOH treatment the cell wall swelled inwardly. accompanied by the decrease in fibre bundle diameter, improving crystalline packing, which also leads to increase in the tensile strength and Young's modulus (Fig. 7 and 8). Thus, layers are swollen and crystalline packing is improved, and no degradation will occur, despite the plasticization of the cell wall. Between 0.16% and 0.24% NaOH treatment further removal of lignin occurred, resulting in further separation of the ultimates thus decreasing the fibre bundle diameter and increasing crystalline packing order. Similar observations have been reported by Preston 1963. This resulted in increased tensile strength and stiffness (Fig. 7 and Fig. 8).

The degradation of the cell wall, progresses inwardly, from the primary wall towards the centre of the fibre in the following order;

Primary wall  $\rightarrow$  S<sub>1</sub>  $\rightarrow$  S<sub>2</sub>  $\rightarrow$  S<sub>3</sub>

Scheme 1. Hypothetical pathway model of progressive swelling of the cell wall following alkalisation

The rate of absorption of NaOH solution and the increasing order of compactness of the crystalline cellulose in the four layers is shown in Scheme 1. The equilibrium at which the absorption will occur at constant concentration of caustic soda will be different from one layer to another and the concentration of caustic soda will have a decreasing effect on the cell wall, becoming less effective progressively towards the centre of the fibre. The mechanical properties of alkalised jute fibre bundles will depend not only on the concentration of caustic soda but also on the sorption time.

### The Effect of Cellulose Content on the Tensile Properties

Figures 9 and 10 show the effect of cellulose content on the tensile strength and Young's modulus of alkalised jute fibre bundles. Following 0.03% NaOH treatment jute fibre bundles exhibited an increase in the cellulose content of about 0.26% that in turn caused increase in tensile strength and Young's modulus of about 11% and 17% respectively. Further increase in cellulose content up to 0.08% NaOH treatment is

expected to decrease non-crystalline materials, particularly wax and pectin, without significantly changing crystalline packing. This organisation of the fine structure of the cell wall will result in increase in the tensile strength and Young's modulus (Fig. 9 and 10). The increase in the cellulose content between 0.08% NaOH and 0.16% NaOH is believed to be due to the increase in the hemicelluloses and lignin present in the primary wall and secondary wall particularly  $S_1$  and  $S_2$  layers. It is also partly due to the increase in the packing order of the crystalline material. The increase in cellulose content between the range of caustic soda treatment mentioned above results in a 48% increase in tensile strength and 15% decrease in Young's modulus. The decrease in the Young's modulus at 0.16% NaOH (Fig. 10) is due to the plasticisation of the secondary wall, which is not accompanied by degradation of the cell wall. Gassan and Bledzki (1999) reported that following alkalisation, release in the strain occurs in the crystalline regions, which results in improved packing order along the longitudinal direction of the fibre with the subsequent increase in mechanical properties.



Figure 9. The effect of cellulose content on the tensile strength of jute fibre bundles with respect to caustic soda treatment



**Figure 10.** The effect of cellulose content on the Young's modulus of jute fibre bundles with respect to caustic soda treatment

#### The Effect of Crystallinity Index on the Tensile Properties

Following alkalisation of the jute fibre bundles both the tensile strength (Fig. 11) and Young's modulus (Fig. 12) show similar pattern of changes in the crystallinity index with respect to changes in the concentration of caustic soda, and the effect these changes has on the tensile strength and Young's modulus of the jute fibre bundles. The crystallinity index was calculated using equation 9. The increase in the crystallinity index between untreated and 0.03% NaOH treated jute fibre bundles are caused by a decrease in non-crystalline materials, particularly wax and pectin. The 0.03% NaOH concentration is not strong enough to cause significant swelling of the cell wall. Further increase in caustic soda treatment of up to 0.08% NaOH would result in further decrease in non-crystalline materials, leaving the component  $I_{(002)}$  in equation 9 unchanged, while at the same time causing reduction in the fire bundle diameter. Crystalline packing in the primary wall will also occur, resulting in an increase in the tensile strength and Young's modulus.

The decrease in the crystallinity index between 0.03% and 0.08% NaOH treatment is attributed to the degradation of the cellulosic materials in the primary wall, whereas an increase in the tensile strength and Young's modulus between this range of caustic soda concentrations indicates the improvement in the packing order of the crystalline materials particularly in the S<sub>1</sub> layer. The increase in the crystallinity index between 0.08% and 0.24% NaOH treatment is due to the decrease in the amorphous materials including hemicelluloses, thus resulting in the increase in the tensile strength

and Young's modulus, although a decrease in the Young's modulus at 0.16% NaOH treatment indicates that the cell wall has undergone plasticisation. The decrease in the tensile strength (Fig. 11) and Young's modulus (Fig. 12) between 0.24% and 0.32% NaOH treatment is associated with plasticisation and degradation of the cell wall.



Figure 11. The dependence of tensile strength of jute fibre bundles on the crystallinity index with respect to caustic soda treatment



**Figure 12.** The dependence of Young's modulus of jute fibre bundles on the crystallinity index with respect to caustic soda treatment

#### The Effect of the Micro-Fibril Angle on the Tensile Properties

The initial postulated stiffness model of the micro-fibril inclined at an angle  $\theta$  to the fibre major axis was presented in equation 7. The general equation for the stiffness of plant fibres has been fully explained in Mwaikambo and Ansell (2006a). For the purpose of determining the micro-fibril angle of jute fibre bundles, equation 8 is modified to obtain equation 11. The micro fibrils are arranged in fabric-like layers known as lamellae, which exhibit alternately either a Z or S winding direction (Morton and Hearle 1975). Using equation 11 an estimate of the micro-fibril angle  $\theta$  was determined,

$$\theta = \cos^{-1} \sqrt{\left(\frac{E_{f} - (1 - V_{s})E_{L}}{V_{s}E_{s}}\right)}$$
(11)

where,  $E_f$ ,  $V_s$ ,  $E_s$  and  $E_L$  are the elastic moduli of fibres, volume fraction, and stiffness of microfibrils, and the stiffness value for non-cellulose materials, respectively. Only the micro-fibril angle of untreated fibres was determined (16.9°), and this is shown in Table 1. In order to evaluate the elastic moduli of the micro-fibrils, it was assumed that following alkalisation (a) the micro-fibril angle and (b) the stiffness of the non-cellulose material remain the same. It is worth noting that caustic soda does not degrade lignin, hence validating the second assumption. Whilst the first assumption may not strictly be valid, combined with the second assumption, acceptable results for the stiffness of the fibre and cellulose content in untreated and alkali treated jute fibre bundles are obtained and are shown in Table 1.

Table 1.	Estimates of the Micro-Fibril Angles of Untreated Jute	Fibre	as a
Function	of Cellulose Content (Z)		

	<b>\ \ \</b>		
Fibre type	Jute		
Wz	0.84		
${\rm E_{z}}$ (GPa)	20.47		
$E_{\rm f}$ (GPa)	17.07		
θ (°)	16.9		

Results in Table 1 indicate that the stiffness of the fibre  $(E_f)$  was approximately 17% lower than the stiffness of the micro-fibrils (cellulose). The decrease in the stiffness of the fibre from that of the micro-fibrils is the result of resolving the forces with respect to the angle of inclination to the longitudinal axis of the fibre and the presence of the non-cellulose materials mainly lignin. Table 1 also shows the micro-fibril angle (16.9°), which is within and close to the values reported in literature (Morton and Hearle 1975; Perry 1975). The variation in the micro-fibril angle between experimental and literature value is also attributed to differences in the environmental testing conditions.

Using the stiffness model of plant fibres (eq. 8) and the physical and structural properties of jute fibre bundles, including the literature values of the stiffness of lignin, 3.0 and 3.4 GPa, reported by Kulkarni et al. (1983)] and McLaughlin and Tait (1980)

respectively, estimates of the elastic moduli of the micro-fibrils ( $E_S$ ) were determined as a function of firstly crystallinity index ( $E_I$ ) and secondly cellulose content ( $E_Z$ ), and are shown in Table 2. The stiffness of the micro-fibrils, obtained using the cellulose content  $E_S$  and the stiffness of the micro-fibrils obtained using the crystallinity index  $E_I$  were obtained by substituting values of  $V_I$ ,  $E_f$ ,  $E_I$ ,  $V_S$ , respectively, and the calculated  $\theta$  and  $E_L$  in equations 12 and 13.

$$E_{s} = \left[\frac{E_{f} - (1 - V_{s})E_{L}}{V_{s} \cos^{2}\theta}\right]$$
(12)

$$E_{I} = \left[\frac{E_{f} \cdot (1 - V_{I})E_{L}}{V_{I} \cos^{2}\theta}\right]$$
(13)

Equations 12 and 13 were developed and used to determine the stiffness of the micro-fibrils with respect to the cellulose content and crystallinity index of the jute fibre. Figure 13 was used to illustrate the difference in the stiffness with respect to the cellulose content (micro-fibril content) and crystallinity index. It was observed that untreated jute fibre exhibits higher stiffness using the crystallinity index than using the cellulose content. Following alkalisation between 0.03% and 0.08% NaOH concentration, the stiffness obtained using the crystallinity index and cellulose content (micro-fibrils) increases. The decrease in  $E_I$  and  $E_S$  is due to the plasticization of the cell wall between 0.08% and 0.16% NaOH treatment. The slight increase in stiffness at 0.24% NaOH treatment is the result of the densification of the crystallites and the decrease in stiffness beyond 0.24% NaOH treatment is due to the degradation of the cell wall caused by the high concentration of caustic soda. Table 2 shows that the elastic modulus (E<sub>1</sub>) values of the crystalline material determined using the crystallinity index (eq. 13) are higher than values of elastic modulus  $(E_s)$  determined using the micro-fibril or cellulose content (volume fraction of cellulose) (eq. 12). This further demonstrates that crystallinity index is a better measure of the plant fibre's stiffness than cellulose content.

Treatment	0	0.03	0.08	0.16	0.24	0.32
(%NaOH)						
E <sub>I</sub> (GPa)	24.21	26.25	33.47	27.03	33.28	14.66
E <sub>z</sub> (GPa)	20.47	23.86	28.68	23.55	30.76	13.34

**Table 2.** The Estimated Elastic Moduli of the Micro-Fibrils of Jute Fibre Bundles

 Calculated as a Function of Crystallinity Index (I) or Cellulose Content (Z)

The results also show that the lower stiffness values obtained using the cellulose content is a result of the presence of tiny air pockets, as reported in earlier findings by Mwaikambo and Ansell (2001), and the lignin content and hemicelluloses, which reduce stiffness. The stiffness values obtained in Table 2 are close to stiffness values obtained by McLaughlin and Tait (1980) and Kulkarni et al. (1983) using bowstring hemp

(*Sansevieria metallica*) and banana (*Musa sepintum*) fibres, respectively. Figure 13 shows the Young's moduli of the micro-fibrils determined using the cellulose content and the crystallinity index of untreated and caustic soda treated jute fibres.



Figure 13. The stiffness of micro-fibrils of jute fibre bundles vs caustic soda treatment with respect to cellulose content and crystallinity index

The moduli  $E_1$  and  $E_s$  reached a maximum at approximately 0.19% NaOH, with insignificant variation when the stiffness values were approximated to one decimal place. The tensile strength versus Young's modulus of jute fibre bundles has been plotted and is shown in Fig. 14. Following alkalisation between untreated and 0.03% NaOH treatment the scatter of the points was almost evenly distributed because within this range of caustic soda concentration the S<sub>1</sub> and S<sub>2</sub> layers were not significantly swollen. However, following 0.08% NaOH treatment the S<sub>1</sub> and S<sub>2</sub> layers were swollen, resulting in plasticisation of the cell wall. Beyond 0.08% NaOH treatment the crystalline materials began to re-order along the fibre main axis and are evenly distributed.

### **Comparison Between Jute, Steel, and Synthetic Fibres**

The specific tensile strength and specific Young's modulus histograms are plotted for untreated and alkali treated jutel fibre bundles and high-performance fibres (Fig. 15 and Fig. 16, respectively) namely steel, E-Glass, Carbon Type I, Carbon Type II, Kevlar 29 and Kevlar 49 fibres. It is observed that the difference between the specific strength of the alkali treated jute fibre bundles and steel is not significant (Fig. 15), whereas the specific strength of the other high performance fibres is much higher than those of untreated and alkali treated jute fibre bundles. The specific stiffness of alkali treated jute fibre bundles is more-or-less the same with that of steel and E-glass fibre. These results show that jute fibre bundles can provide useful replacements for steel and E-glass fibres where specific stiffness is not a critical requirement.



Figure 14. Tensile strength vs Young's modulus of jute fibre bundles







**Figure 16.** A comparison of the specific modulus of jute fibre bundles and E-glass, steel, carbon I, carbon II, Kevlar 29 and Kevlar 49 fibres (numbers appearing on jute fibres are for 0%, 0.03%, 0.08%, 0.16%, and 0.24% NaOH treated fibres)

### Fracture Surface Topography of the Fibre Bundles

Jute fibres have been found to consist of bundles of single ultimate cells, as indicated in the SEM micrographs Figs. 3-5 and also examined in this section. The fibres contain cell wall layers with the lumen space (indicated by arrows) at the centre (Fig. 3-5). Figures 17-22 show SEM micrographs of jute fibre bundles fractured in tension. Figure 17 shows a brittle fracture of untreated jute fibre bundle. Fibre pull-out is observed in the outer part of the bundle (small arrows), which indicates lack of sufficient binding material (mostly lignin and pectin). The fracture failures at the centre of the alkalised fibre bundle exhibit no fibre pull-out (long arrows), indicating the presence of sufficient binding materials (long arrows). Fractured fibre bundles also exhibit insignificant fibril splitting and fibre buckling. Furthermore, the failure of the jute fibre bundle is not at the same stress level, indicating the presence of cell wall defects along the fibre, which then develop stress intensities leading to failure.

Figure 18 shows brittle fracture of 0.03% NaOH treated jute fibre bundle. The fracture mechanism is similar to that exhibited by the untreated sisal fibre bundle (Fig. 17). Fibre splitting and fibre pull-out are in the form of layers and appear to have occurred to the middle layers (S<sub>1</sub>, S<sub>2</sub> and S<sub>3</sub> layers), which implies that the primary layer has been degraded following alkalisation and that the secondary layers are least affected by the 0.03% NaOH treatment. Figure 19 shows brittle fracture of 0.08% NaOH treated jute fibre bundle. The fracture was more-or-less the same across the transverse section of the fibre bundle. Pull-out of the ultimate fibres was not significant. However, the pull-out in one of the ultimates was on the outer layer and it was due to plasticization. Ultimate fibres around the outer part of the bundle also exhibited buckling following fracture, and insignificant splitting of the fibrils was observed. The minimal splitting of the fibres into fibrils is an indication of minimal degradation but higher plasticization.

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Figure 20 shows a fracture surface of 0.16% NaOH treated jute fibre bundle. Fibre pull-out and buckling are not observed, and the cells break at around the same level indicating that the packing order of the crystalline cellulose is evenly distributed across and along the fibre. This also indicates that 0.16% NaOH treatment has degraded the binding material and penetrated the secondary wall. This re-organisation of the crystalline region will have significant increase in the tensile strength and Young's modulus of the jute fibres. Figure 21 shows the 0.24% NaOH treated jute fibre bundle fractured in tension. The ultimate fibres showed significant swelling of the cell wall and have fractured at the same level. There was no fibril splitting, buckling was observed, and the fibre cells showed no lumen. The absence of split fibrils and lumen is an indication of complete swelling of the cell wall. Figure 22 shows a 0.32% NaOH treated jute fibre bundle fractured in tension. The fibres size in transverse direction has been reduced and showed buckling. The fracture was at different level, which shows that the cell wall has been degraded.



**Figure 17.** SEM micrographs of untreated jute fibre bundles fractured in tension



**Figure 18.** SEM micrographs of 0.03% NaOH treated jute fibre bundles fractured in tension



**Figure 19.** SEM micrographs of 0.08% NaOH treated jute fibre bundles fractured in tension



**Figure 20.** SEM micrographs of 0.16% NaOH treated jute fibre bundles fractured in tension



**Figure 21.** SEM micrographs of 0.24% NaOH treated jute fibre bundles fractured in tension



**Figure 22.** SEM micrographs of 0.32% NaOH treated jute fibre bundles fractured in

#### CONCLUSIONS

In general the stiffness of jute fibre bundles showed an increasing trend following treatment using caustic soda. The increase was optimised at 0.24% NaOH (by weight), beyond which the tensile strength and stiffness decreased. The tensile strength and stiffness were directly related to the amount of cellulose content and the crystallinity index, and the crystallinity index was an accurate measure of stiffness of jute fibre bundles than cellulose content. The swelling of the jute fibre cell wall following alkalisation varied within the fibre, and the variation is believed to be due to the densification of the crystalline regions. The tensile properties and the physical changes obtained in this study following alkalisation showed that the effect of caustic soda to the plant cell wall varied across the fibre. It also showed that equilibrium was not attained at the caustic soda concentrations applied due to degradation effect of caustic soda to the plant cell wall, while maintaining the morphological structure. The increase in the mechanical properties of caustic soda treated jute fibre bundles indicates its potential application as reinforcement for polymeric materials for composite manufacture. The results in this research work have also shown that jute fibre is a potential alternative to fossil-based reinforcements.

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