The effect of chemical treatment on the properties of hemp, sisal, jute and kapok fibres for composite reinforcement

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ABSTRACT

Two chemical treatments were applied to hemp, sisal, jute and kapok natural fibres to create better fibre to resin bonding in natural composite materials. The natural fibres have been treated with varying concentrations of caustic soda with the objective of removing surface impurities and developing fine structure modifications in the process of mercerisation. The same fibres were also acetylated with and without an acid catalyst to graft acetyl groups onto the cellulose structure, in order to reduce the hydrophilic tendency of the fibres and enhance weather resistance.

Four characterisation techniques, namely XRD, DSC, FT-IR and SEM, were used to elucidate the effect of the chemical treatment on the fibres. After treatment the surface topography of hemp, sisal and jute fibres is clean and rough. The surface of kapok fibres is apparently not affected by the chemical treatments. X-ray diffraction shows a slight initial improvement in the crystallinity of the fibres at low sodium hydroxide concentration. However, high caustic soda concentrations lower the fibre crystallinity. Thermal analysis of the fibres also indicates reductions in crystallinity with increased caustic soda concentrations and that grafting of the acetyl groups is optimised at elevated temperatures. Mercerisation and acetylation have successfully modified the structure of natural fibres and these modifications will most likely improved the performance of natural fibre composites by promoting better fibre to resin bonding.

Key words: natural fibres, sisal, jute, hemp, kapok, chemical treatment, mercerisation, acetylation, characterisation, XRD, DSC, FT-IR and SEM.

1. INTRODUCTION

1.1. Plant fibres as reinforcement for composites

The increase in the application of plant fibres as reinforcement for polymeric substrates has been stimulated by the environmental cost of manufacturing energy-intensive, synthetic fibres such as glass, carbon and kevlar. However, whereas synthetic fibres can be produced with engineered properties to suit particular applications this is not the case with naturally occurring plant fibres. Properties of the cellulose fibres depend mainly on the nature of the plant, locality in which it is grown, age of the plant and extraction method used. For instance, sisal is a hard leaf fibre but jute and hemp are both bast fibres and are generally referred to as 'soft' fibres to distinguish them from the hard leaf fibres. Both leaf and bast fibres are multi-cellular with very small individual cells bonded together (Preston, 1963; Hearle, 1963 and Hegbom, 1990). Kapok is a single cell seed fibre with a wide air lumen.

The mechanical properties of plant fibres are largely related to the amount of cellulose, which is closely associated with the crystallinity of the fibre and the micro-fibril angle with respect to the main fibre axis (Sreekala et al, 1997). Fibres with high crystallinity and/or cellulose content have been found to possess superior mechanical properties. Sisal fibres with a cellulose content of 67% and micro-fibril angle of 10-22° have a tensile strength and modulus of elasticity of 530 MPa and 9-22 GPa respectively. On the other hand, coir fibre with a cellulose content of 43% and micro-fibril angle of 30-49° is reported to have a tensile strength and modulus of elasticity of 106 MPa and 3 GPa respectively (Hegbom et al, 1990, Hearle et al, 1963 and Preston, 1963). This variation in the mechanical properties with increased microfibril angle plays an important role in determining the mechanical properties of fibre reinforced composites. In addition it is necessary to optimise fibre alignment parallel with the direction of applied force to maximise tensile properties.

1.2. Fibre to matrix interfaces in natural fibre-reinforced composites

The performance and stability of fibre-reinforced composite materials depends on the development of coherent interfacial bonding between fibre and matrix. In natural fibre-reinforced composites there is a lack of good interfacial adhesion between the hydrophilic cellulose fibres and the hydrophobic resins due to their inherent incompatibility. Short, cellulose-based fibres will also tend to agglomerate making their use in reinforced composites less attractive. The presence of waxy substances on fibre surface contributes immensely to ineffective fibre to resin bonding and poor surface wetting is observed. Also the presence of free water and hydroxyl groups, especially in the amorphous regions, worsens the ability of plant fibres to develop adhesive characteristics with most binder materials. High water and moisture absorption of the cellulose fibres causes swelling and plasticising effect resulting in dimensional instability and poor mechanical properties. Plant fibres are also prone to micro - biological attack leading to weak fibres and reduction in their life span (Bisanda and Ansell, 1992).

Fibres with high cellulose content have also been found to contain a high crystallite content. These are the aggregates of cellulose blocks held together closely by the strong intra-molecular hydrogen bonds which large molecules, for example dyes, are not able to penetrate unless the cell wall is swollen. Fibres are, therefore, usually subjected to treatment such as mercerisation and acetylation, with or without heat, to first bulk or swell the cell wall to enable large chemical molecules to penetrate the crystalline regions.

1.3. Chemical treatment of natural fibres – mercerisation

Natural fibres are chemically treated in order to remove lignin-containing materials such as pectin, waxy substances and natural oils covering the external surface of the fibre cell wall. This reveals the fibrils and gives a rough surface topography to the fibre. Sodium hydroxide (NaOH) is the most commonly used chemical for bleaching and/or cleaning the surface of plant fibres. It also changes the fine structure of the native cellulose I to cellulose II by a process known as mercerisation (Johnson and Shenouda, 1979). The reaction of sodium hydroxide with cellulose is thought to be as follows.

$$\text{Cell - OH} + \text{NaOH} \longrightarrow \text{Cell - O}^{-}\text{Na}^{+} + \text{H}_2\text{O} + [\text{ surface impurities }] \qquad (1)$$

It is worth pointing out that mercerisation de-polymerises the native cellulose I molecular structure producing short length crystallites. However, there seems to be varying interpretations of the term 'mercerisation'. The standard definition for mercerisation proposed by ASTM D1695 is "the process of subjecting a vegetable fibre to the action of a fairly concentrated aqueous solution of a strong base so as to produce great swelling with resultant changes in the fine structure, dimension, morphology and mechanical properties". ASTM D123-83a defines mercerised yarn as "a cotton yarn which has been treated with a solution of sodium hydroxide under conditions of concentration and temperature which

effect a permanent or irreversible swelling of the cellulose". In both definitions neither the alkali concentration nor the treatment temperature are mentioned.

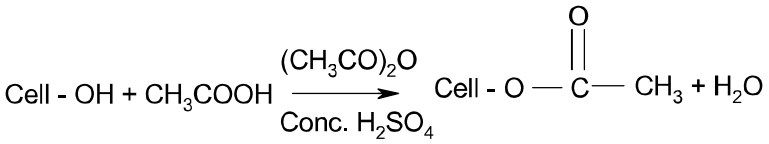
Zeronian (1985) proposes another definition of mercerisation, which is suitable for basic research and is more specific. Mercerised cellulose is "a sample of cellulose which has been treated with a solution of an alkali metal hydroxide of sufficient strength to cause essentially complete conversion of the crystal structure from cellulose I to II". It is reported that residual traces of cellulose I are found even when the strength of the alkali used in the mercerisation treatment is considered optimum for conversion (Zeronian, 1985). Atkins (1979) reports that mercerisation without tension allows total conversion of cellulose I to cellulose II to take place. However the application of tension to maintain the even distribution of crystallites only allows partial conversion.

Owolabi *et al* (1985) used 50%NaOH on coconut fibres while Sreekala *et al* (1997) and Geethamma *et al* (1995) used 5%NaOH to remove surface impurities on oil palm fibres and short coir fibres respectively. Bisanda and Ansell (1992) applied a concentration of 0.5N NaOH on sisal fibre, which resulted in improvement in the mechanical properties of the sisal fibre. However, Murkhejee *et al* (1993) found that the use of more than 1% NaOH on cellulose fibres weakens the fibres resulting in poorer mechanical properties. This last finding appears to be in perfect agreement with the commonly held principle of mercerisation, which states that caustic soda increases the amount of amorphous regions at the expense of crystallinity.

It is reported that mercerisation, both slack and with tension increases the strength uniformity along the fibre length. This is attributed to an increase in strength at the weakest point in the fibre (Shenouda, 1979 and Hearle, 1963). Mercerisation also improves accessibility of reactive sites to dyes and binding chemicals bringing about crystalline modification which involves fibril swelling and sometimes improves the crystalline packing order which has the advantage of providing more access to penetrating chemicals. The presence of reactive sites and fibril swelling are prerequisite factors to resin cross-linking inside the fibre. Cellulose-based fibres absorb moisture causing both reversible and irreversible swelling. In composite products this can result in undesirable dimensional changes. To arrest this problem, the reinforcing cellulose fibres are subjected to certain modifications. These processes involves either the stabilisation of the cell wall matrix to restrain swelling or reduction of the hygroscopicity of the cell wall and bulking of the cell wall polymers to maintain the wet volume so that moisture does not cause any additional swelling (Rowell, 1992)

1.4. Chemical treatment of natural fibres – acetylation

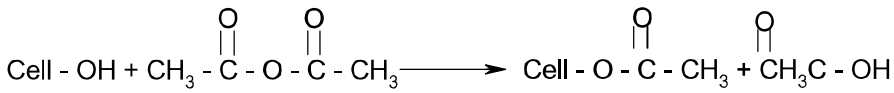
Acetylation, which is another chemical treatment covered in this study, has been extensively applied to wood cellulose to stabilise the cell wall, improving dimensional stability and environmental degradation (Rowell, 1992). The process involves the soaking of the plant fibre in acetic anhydride with or without an acid catalyst as shown in equation 2 and 3 respectively.



(2)

Acetylation with acid catalyst

(3)



Acetylation without acid catalyst

Since acetic acid does not react sufficiently with cellulose, acetic anhydride is preferred instead (Mark et al, 1972 and Moncrieff, 1975). However, because acetic anhydride is not a good swelling agent for cellulose, in order to accelerate the reaction, cellulose materials are first soaked in acetic acid and subsequently treated with acetic anhydride at higher temperatures for a period of between 1 and 3 hours. The rate of reaction is much faster with fibres that have not been mercerised than with mercerised cellulose fibres. In natural fibre reinforced composite acetylation of the hydroxyl group will swell the plant fibre cell wall, greatly reducing the hygroscopic nature of the cellulose fibre. This will consequently result in dimensional stability of the composites, as any absorbed water will not cause further swelling or shrinkage of the composite material. While the acetylation treatment has long been used on textile goods, it is only recently that research work is being carried out to assess its usefulness in natural fibres for applications in composites (Sreekala *et al*, 1997; Abdul Khalil, 1999).

1.5. Objectives

To properly assess changes at the fibre surface and fine structure due to chemical treatment it is necessary to employ appropriate analytical characterisation methods. A combination of two or more characterisation techniques allows a much more thorough investigation of the effect of chemical treatment on cellulose based fibres. In this study, therefore, four characterisation methods were employed. Wide angle X-ray analysis (WAXS), differential scanning calorimeter (DSC), Fourier transform infrared spectroscopy (FT–IR) and scanning electron microscopy (SEM) have been used to analyse the effect of mercerisation and acetylation on the crystallinity and thermal characteristics of four types of natural fibres.

2. EXPERIMENTAL PROCEDURE

2.1. Materials

Sisal and jute fibres used in this work were supplied by the Department of Materials Science and Engineering at the University of Bath. Kapok fibre was obtained from Morogoro in Tanzania whilst hemp was kindly supplied by the Hemcore Company Ltd of United Kingdom. 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% strength, sulphuric acid with 99% strength and glacial acetic acid were supplied as general laboratory reagents. Merck Ltd of England supplied acetic anhydride with density of 1.08 g/cm^3 and boiling temperature of 140 °C. Chemicals were diluted to the concentrations stipulated below.

2.2. 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^3 of water. Assuming the room temperature is 20 ± 2°C, the conditioning chamber with the solution in it generates a relative humidity inside the chamber of approximately 65 ± 2% relative humidity (WIRA, 1973). Fibres for the acetylation treatment were soaked in cold distilled water for 48 hours at 20 ± 2°C to remove any surface impurities that would prevent effective action of the acetyl groups on the cellulose structure before they were conditioned. This process was deemed unnecessary for fibres designated for alkali treatment for reasons which will be explained in the alkali treatment section. Fibre treatments are summarised in Table 1.

2.3. Alkali treatment

Kapok, sisal, jute and hemp were soaked in beakers containing caustic soda concentrations as shown in Table 1 and placed in a water bath controlled at 20 ± 2°C for 48 hours. 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.

Table 1: Specifications of chemical treatment of the fibres.

Treatment		Kapok °C	Sisal °C	Jute °C	Hemp °C
Untreated		-	-	-	-
10% acetic acid (1 hour) + acetic anhydride in sulphuric acid: for 5 min.		20	20	20	20
	1 hour	70	20	70	70
	3 hours	70	70	70	70
10% acetic anhydride for:	1 hour	20	20	20	20
	1 hour	70	70	70	70
	3 hours	70	70	70	70
0.8% NaOH for 48 hours		20	20	20	20
2% NaOH for 48 hours		20	20	20	20
4% NaOH for 48 hours		20	20	20	20
6% NaOH for 48 hours		20	20	20	20
8% NaOH for 48 hours		20	20	20	20
30% NaOH for 48 hours		20	20	20	20
400% (4M) NaOH for 48 hours		20	20	20	20

2.4. Acetylation

One set of the four fibres namely kapok, sisal, jute and hemp were treated in glacial acetic acid (Table 1) for I hour at 20 ± 2°C. It was further treated with acetic anhydride containing concentrated H_2SO_4 as a catalyst for 5 minutes. Fibres were then washed with distilled water and dried. A second set of fibres was treated in acetic anhydride without acid catalyst for one hour at 20 ± 2°C. The fibres were removed and washed with distilled water and dried. The third and fourth sets of fibres were treated in acetic anhydride without acid catalyst in a water bath controlled at 70 °C for 1 hour and three hours respectively. The fibres were then washed with distilled water and dried.

2.5. Differential scanning calorimetry

Untreated and treated fibre samples weighing between 5 and 10 mg were placed in an aluminium capsule, sealed and punctured to allow gases to escape during heating. A Du-Pont DSC 2910 equipped with TA instruments was operated in a dynamic mode with a heating scheme of 30 to 500 °C and heating rate of 10 °C/min. in a nitrogen environment purged at 25 ml/min. The thermograms were analysed for any changes in the thermal behaviour of the fibres.

2.6. X-ray diffraction (WAXS)

Untreated and treated fibres were mixed with a very small amount of a adhesive material Tragacanth BP, soaked in a drop of distilled water and compressed into thin sheets and dried. A wide-angle diffractometer equipped with a scintillation counter and a linear

intensities were recorded between 5° and 60° (2θ -angle range). The crystallinity index (I_c) was determined by using equation 4 below. Where $I_{(002)}$ is the counter reading at peak intensity at a 2θ angle close to 22° and $I_{(am)}$ is the amorphous counter reading at a 2θ angle of about 18°.

$$I_c = \frac{\left(I_{(002)} - I_{(am)}\right)}{I_{(002)}} \text{ x } 100 \qquad (4)$$

2.7. Infrared spectroscopy

Infrared spectra were obtained using a Perkin Elmer FT-IR Spectrometer model PARAGON 1000. About 2 mg of fibre was crushed into small particles in liquid nitrogen. The fibre particles were then mixed with KBr and pressed into a small disc about 1 mm thick.

2.8. Scanning electron microscopy (SEM)

SEM micrographs of fibre surfaces and cross sections of untreated and treated fibres were taken using a scanning electron microscope Model JEOL 6310. Prior to SEM evaluation, the samples were coated with gold by means of a plasma sputtering apparatus.

3. RESULTS AND DISCUSSION

3.1. Differential scanning calorimetry of mercerised fibres

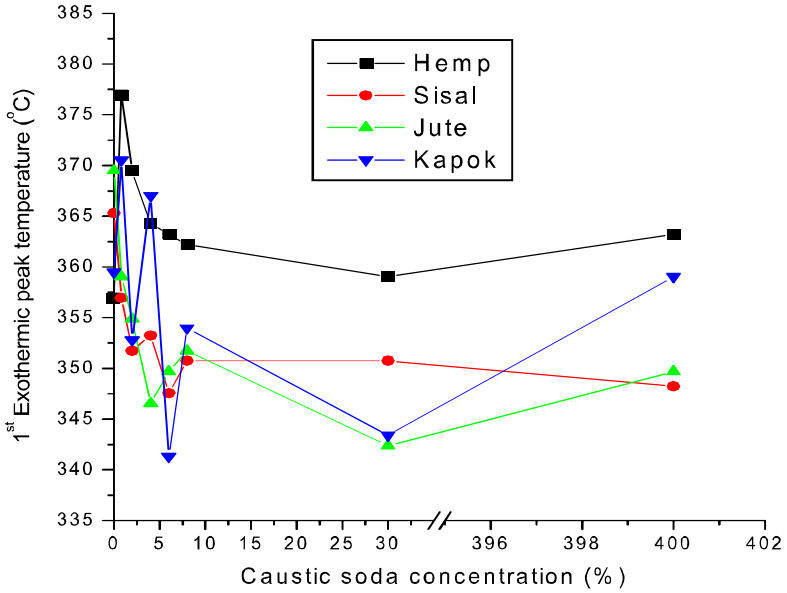


Figure 1: The effect of caustic soda on hemp, sisal, jute and kapok as observed by the DSC method.

DSC analysis enables to identify the chemical activity occurring in the fibre as heat is applied and in this case it was possible to observe one endothermic peak at temperatures between 70-100 and two exothermic peaks at higher temperatures shown in Table 2. The first exothermic peak reflects the stability of the fibres as a function of caustic soda concentration. Figure 1 shows a sharp decreasing trend of the first exothermic peak of temperature as the concentration of the caustic soda is increased from 0.8% to 6%. The peak temperature then slowly decreases up to 30% NaOH beyond which a slight increase is observed. The information is also summarised in Table 2.

Table 2. Crystallinity and first exothermic peak temperature as a function of concentration of alkali treatment (second exothermic peak temperature in brackets).

NaOH (%)	Hemp		Sisal	
	Crystallinity (%)	Exotherm peaks (ºC)	Crystallinity (%)	Exotherm peaks (ºC)
0	87.87	357.00 (390.35)	70.9	365.31 (444.63)
0.8	88.79	377.03 (389.31)	71.04	356.96 (398.70)
2	88.75	369.50 (386.18)	68.93	351.75 (412.26)
4	87.61	364.27 (389.31)	74.66	353.24 (387.04)
6	86.54	363.22 (382.00)	79.30	347.57 (394.52)
8	87.70	362.18 (397.65)	75.11	350.74 (393.30)
30	89.77	359.05 (389.31)	78.77	350.74 (396.61)
4M	81.34	363.22 (414.35)	73.51	348.23 (374.52)

NaOH (%)	Jute		Kapok	
	Crystallinity (%)	Exotherm peaks (ºC)	Crystallinity (%)	Exotherm peaks (ºC)
0	78.47	369.48 (417.48)	45.75	359.50 (402.87)
0.8	76.61	359.05 (417.48)	46.31	370.53 (434.17)
2	83.10	354.88 (395.57)	49.5	352.79 (402.00)
4	83.06	346.55 (375.77)	48.73	367.00 (406.00)
6	82.37	349.66 -	48.02	341.31 (416.43)
8	82.50	351.75 (444.60)	58.42	353.96 (404.96)
30	82.50	342.36 (391.39)	62.41	343.40 (409.13)
4M	78.32	349.66 -	53.74	359.05 (385.13)

Comparing the untreated fibres, hemp is the least resistant to thermal treatment. However, once treated with caustic soda hemp fibre gives higher thermal resistance than the rest of the alkali treated fibres except at 4% NaOH concentration where Kapok fibre gives higher thermal resistance. Sisal and jute show a drastic decrease in thermal resistance when subjected to concentrations of 0.8% and 4% NaOH and then the resistance rises slightly between 4 and 8% NaOH concentration after which it

drops. With the exception of sisal, the thermal resistance of other fibres rises above 30% NaOH concentration.

3.2. Differential scanning calorimetry of acetylated fibres

The thermal characteristics of acetylated hemp, sisal, jute and kapok fibres are shown in Figure 2.

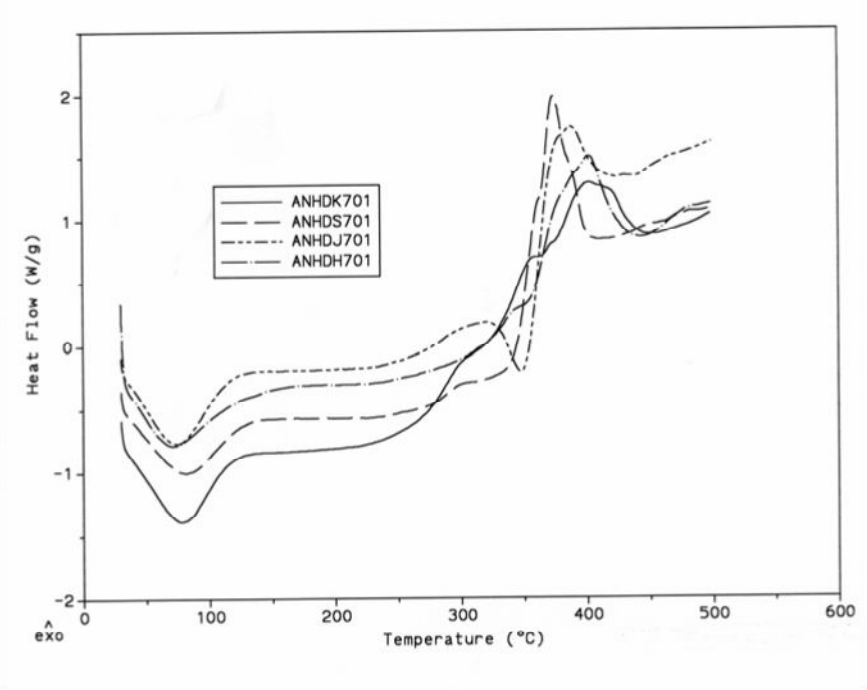


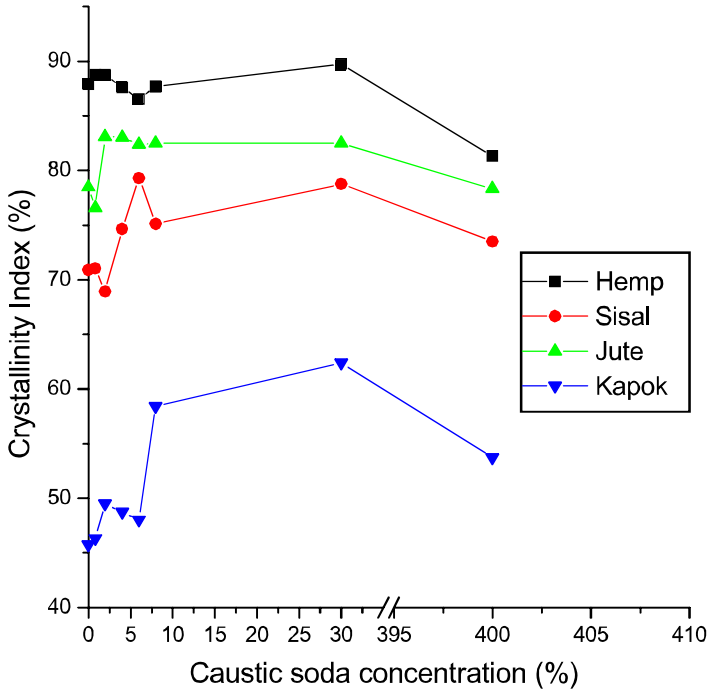
Figure 2: The effect of acetylation on hemp (ANHDH701), sisal (ANHDS701), jute (ANHDJ701) and kapok (ANHDK701) as observed by the DSC method.

All the fibres except kapok show an endothermic peak at around 80 ºC, which is due to water, desorption. The breaking down of the acetyl group causes the second endothermic peak observed in jute fibre. Beyond this peak nearly all the fibres show an exothermic peak between 380 ºC and 400 ºC while kapok fibre has two minor exothermic peaks between 300 ºC and 350 ºC and one major peak at around 400 ºC. On application of heat jute fibre is seen to be less thermally stable than the rest of the fibres and this is more apparent at elevated temperatures. The exothermic peak seen in jute, hemp and sisal is more regular in acetylated fibres.

3.3. X-ray diffraction of mercerised fibres

As the concentration of caustic soda increases, X-ray diffraction results for mercerised fibres (Figure 3 and Table 2) show an overall increase in crystallinity index (Equation 4) for sisal, jute and kapok fibre while that of hemp fibre decreases. Hemp fibre has the highest crystallinity index at any of the caustic soda concentrations followed by jute, sisal and kapok respectively. The effect of mercerisation on the fibres was not the same at each of the applied caustic soda concentrations. For example, at 0.8% NaOH, jute fibre has a lower crystallinity index than the untreated jute fibre whereas a slight increase in crystallinity is observed in hemp, sisal and kapok at the same alkali strength in contrast with the untreated fibres. At 2% NaOH sisal, hemp and kapok show increases in crystallinity index while jute fibre shows a decrease in the crystallinity index. Hemp and kapok fibres have their highest crystallinity index at 30% NaOH, sisal at 6% NaOH, and jute at 4% NaOH.

Figure 3: Crystallinity index versus caustic soda concentration for untreated and treated hemp, sisal,



jute and kapok fibres measured by WAXS.

3.4. FT-IR analysis of acetylated fibres

The grafting of acetyl groups to fibre cell walls is of considerable importance to this work. FT-IR reveals the extent of grafting. The characteristic peaks observed are summarised in Table 3. The peak observed at 3440 cm^{-1} in untreated fibres indicates the presence of intermolecular hydrogen bonding and tends to shift to higher absorbency values in acetylated fibres, e.g. 3480 cm^{-1} in sisal, jute and hemp fibres but remains unchanged at around 3400 cm^{-1} in kapok fibre. The increase in peak intensity at 1743 cm^{-1} in sisal, jute and hemp and kapok fibres is due to the bonded acetyl group. The peak at 1638 cm^{-1} in hemp fibre is due to removal of unsaturated C=C stretching present in traces of oils.

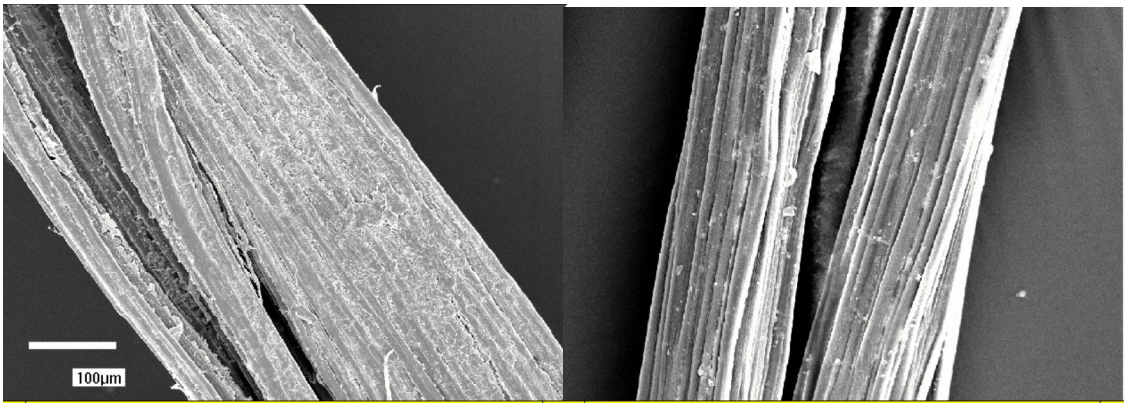
Similar observations have been reported in earlier work on acetyaltion of the wood cell wall (Clemsons *et al*, 1992; Mallari *et al*, 1989 and Rowell *et al*, 1994). The increase in absorbency in the region between 1000 – 1500 cm^{-1} bands shows the increase in O-H stretching, indicating that there has been a reduction in the number of hydroxyl groups at the 3400 to 3500 cm^{-1} band.

Table 3: Infrared transmittance peaks (cm^{-1}) of acetylated fibres relative to untreated fibres.

Bond type	Hemp	Sisal	Jute	Kapok
Intermolecular hydrogen bonding, C-H stretching	3448	3423	3471	3406
C-H stretching	2916	2920	2920	2920
Carboxylic anhydride	1638	1740	1743	1740
C-H bending	1383	1384	1384	1374
C-H bending	-	1250	1248	1244
C-C stretching	1059	1509	1059	1057

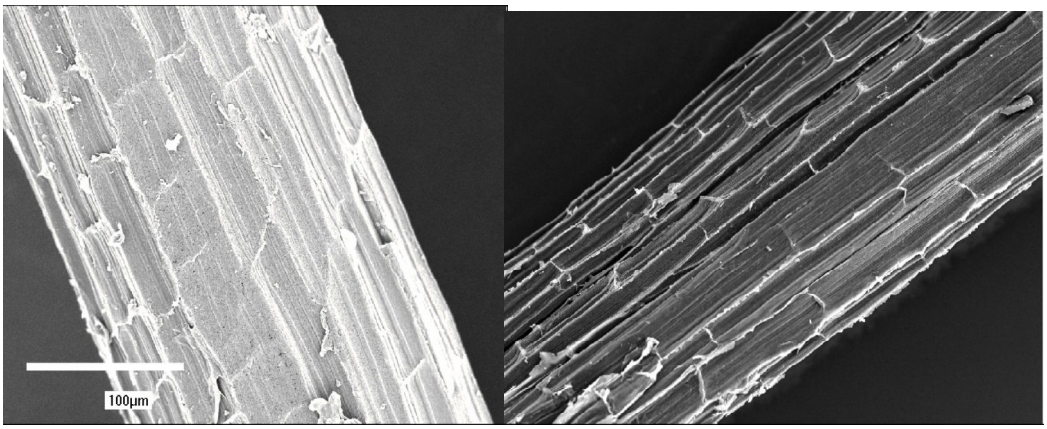
3.5. Scanning electron microscopy of mercerised fibres

Following mercerisation the surface topography of jute, sisal and hemp fibres is rougher than before treatment (Figures 4, 5 and 6).



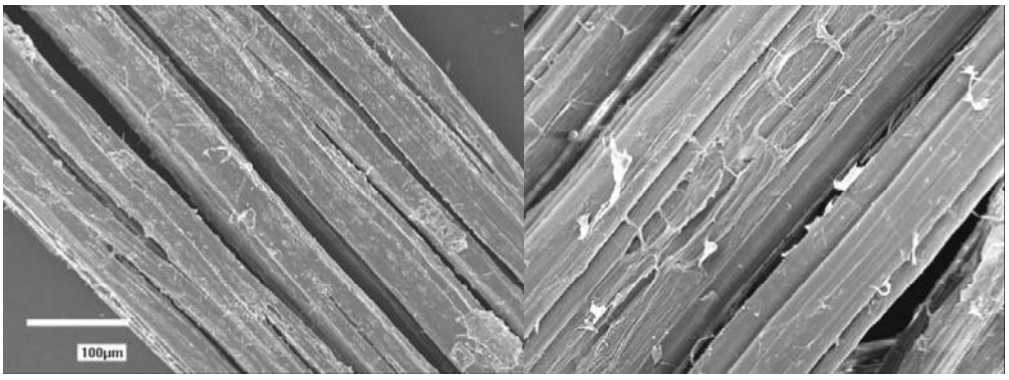
(a) (b)

Figure 4 (a) Untreated jute fibre and (b) alkali treated jute fibre



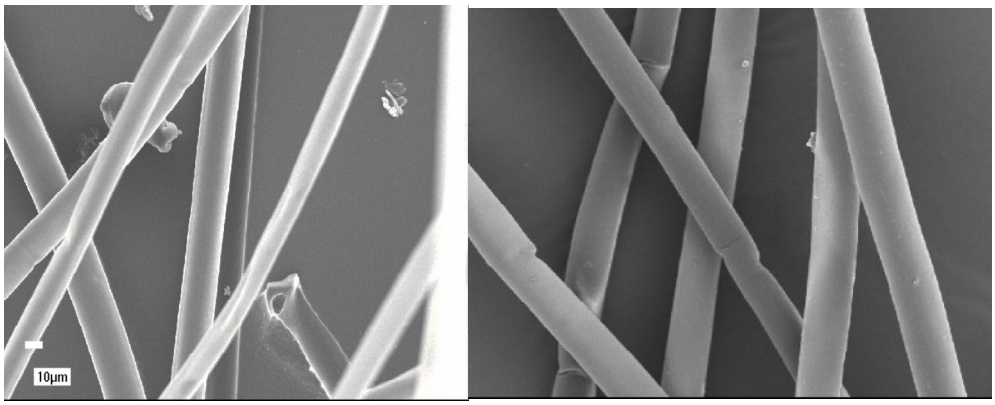
(i) (ii)

Figure 5. Untreated sisal fibre (a) and alkali treated sisal fibre (b).



(a) (b)

Figure 6. (a) Untreated hemp fibre and (b) alkali treated hemp fibre.



(a) (b)

Figure 7. Untreated kapok fibre (a) and alkali treated kapok fibre (b).

Sisal, hemp and jute comprise bundles of individual cells that have been bound together by lignin-rich, weak inter-molecular bonds. Sisal fibres are discontinuous, comprising short lengths joined together to end, whereas hemp and jute fibres are continuous. Figure 4(b) show ridges on the surface of clean jute fibre after alkali treatment. Hemp fibre shows partly separated individual cells before mercerisation, Figure 6(a). Alkali treated hemp fibre looks cleaner and fibre bundles are more separated, with a highly serrated surface. The surface of kapok fibres appears to be unaffected by mercerisation (Figure 7).

4. DISCUSSION

4.1 Differential scanning calorimetry

Caustic soda treatment of plant fibres has, therefore, dual advantages; it removes the fibre surface impurities (Figures 4 to 7) and more importantly, modifies the crystallites of the cellulose (Figure 3). The process also swells the fibre to enhance the crystallite order and increase chemical uptake. The DSC technique is reported (Nguyen *et al*, 1981) to be a very useful tool to determine the drop in crystallinity and decomposition of plant fibre cellulose. The results obtained in this work using the first exothermic DSC peak (removal of moisture) corresponds well with the results obtained using the second and stronger endothermic peak (reduction in the crystallinity) to assess the thermal degradation of crystallites in plant fibres. Shenouda, 1979 and Nguyen *et al*, 1981 have made similar observations with respect to the second peak.

The sharp decrease in the decomposition temperature of the mercerised plant fibres between 0.8% and 8% NaOH concentration is an indication of the increase in amorphous cellulose, known to have poor thermal resistance, and a decrease in cellulose crystallite length. This view is shared by several researches (Aboul-Fadl et al, 1985, Bisanda and Ansell, 1992) It is observed in Figure 1 that more than 8% caustic soda renders the converted cellulose structure slightly stable to thermal degradation (Figure 1). Alkali treated hemp fibre is more crystalline than sisal, jute and kapok (Table 2 and Figure 3) and was found to be more stable to thermal degradation as measured by the DSC.

Some researchers report that mercerised plant fibres used as reinforcement in the manufacture of composites improve mechanical properties in comparison with non-mercerised fibres (Marcovich *et al*, 1998, Bisanda and Ansell, 1992). Mwaikambo and Bisanda, 1999, found that the application of 5% sodium hydroxide to cotton/kapok fabric for reinforcement of unsaturated polyester resin increased tensile strength but decreased modulus of elasticty and impact strength. Aboul-Fadil *et al*, 1985, found that there was a decrease in the breaking strength and tenacity of mercerised Pima S-5, Giza 76, and Giza 77 cotton species while mercerised Giza 75, Giza 80 Dendara, Deltapine Smooth Leaf produced

higher breaking and tenacity values. This implies that the mercerisation of plant fibres can have different effects on the mechanical properties of fibres and also composite materials reinforced with these fibres.

4.2. X-ray diffraction

X-ray results for mercerisation which show an overall increase in the 'crystallinity' index indicate improvement in the order of the crystallites as the cell wall thickens upon alkali treatment. Alkali treatment is reported to reduce the proportion of crystalline material present in plant fibres, as observed by several researchers (Aboul-Fadl, 1985, Hearle, 1975, and Hearle 1963. It is therefore difficult to reconcile the results of this work. However, since mercerisation with and without tension increases the crystallite packing order, it is therefore logical to deduce that the order of the crystallites improves rather than the crystallinity increasing.

4.3. FT-IR analysis

The FT-IR results Table 3 indicates that some chemical reactions occurring during acetylation of the fibres and the presence of a peak in all the fibres at 1740 cm^{-1} is caused by the reaction of the ester groups present at 1734 cm^{-1} in the untreated fibres with the acetyl groups observed at the former peak. Similarly, the reduction of the intermolecular hydrogen bonding between 3406 and 3471 cm^{-1} confirms the grafting of the acetyl groups on the cellulose structure thus replacing the hydroxyl groups. Sreekala, 1997 and Hill *et al*, 1998 studied the acetylation of coir fibre and found similar results. Acetylation of plant fibres reduces the hygroscopic nature of the cell wall and the incorporation of acetylated fibres into plastics enhances weather resistance, thermal resistance and dimensional stability of the composites (Moncrieff, 1975, Rowell, 1992 and Obataya, 1999).

5. Conclusion

- Mercerisation of plant fibres effectively changes the surface topography of the fibres and their crystallographic structure. However care must be exercised in selecting the concentration of caustic soda for mercerisation as results show that some fibres at certain NaOH concentration have reduced thermal resistance as elucidated by the DSC method.

- It is believed that the increase in the crystallinity index obtained by X-ray diffraction is in actual fact an increase of the order of the crystallite packing rather than in increase in the intrinsic crystallinity.

- It is essential therefore to use several complementary techniques when studying the fine structure of natural fibres to confirm trends and that the application of the DSC technique probably gives a better analysis of the fine structure of the plant fibres than the X-ray method alone.

- The removal of surface impurities on plant fibres may be an advantage for fibre to matrix adhesion as it may facilitate both mechanical interlocking and the bonding reaction due to the exposure of the hydroxyl groups to chemicals such as resins and dyes.

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