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RESEARCH PAPER

Structural characterization of coconut tree leaf sheath fiber reinforcement

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Abstract: The coconut palm tree leaf sheath fibers were analyzed by FTIR spectral analysis, Chemical, X-ray and thermo gravimetric methods to assess their suitability as reinforcements in the preparation of green composites. The morphology of the untreated and alkali treated fibers was studied by scanning electron microscopic method. The FTIR and chemical analyses indicated lowering of hemi-cellulose content by alkali treatment of the fibers. The X-ray diffraction revealed an increase in crystallinity of the fibers on alkali treatment. The thermal stability of the fibers was found to increase slightly by alkali treatment. The tensile properties of these fibers increased on alkali treatment. The mechanical and other physical properties indicated that these fibers were suitable as reinforcements for making the green composites.

Keywords: coconut leaf sheath fibers; chemical analysis; crystallinity; mechanical properties; scanning electron microscopy

Introduction

Since the last decade of the 20th century, the usage of natural products (wood, fibers and agro waste) as reinforcements in composites has increased dramatically. Environmental concern is one driving force that led to the consideration of biodegradable ligno cellulose fibers for this purpose (Mohanty et al. 2000). Natural fibers have some advantages over the man-made fibers, including low cost, lightweight, renewable character, high specific strength and modulus, and availability in a variety of forms throughout the world (Mohanty et al. 2000; Rajulu et al. 2002; Maheswari et al. 2008). The reactive surface and the possibility to generate energy, without residue, after burning at the end of their life-cycle, motivate their association with organic polymers in the preparation of green composites. Hence, many efforts are being made to make natural fiber reinforcement thermoplastic and thermosetting composites (Taha and Ziegmann 2006; Li et al. 2004; Teramoto et al. 2004; Sydenstricker et al. 2003; Mohanty and Nayak 2006; Bledzki et al. 1996; Rajulu et al. 2007).

Coconut tree is native to coastal areas of Southeast Asia (Malaysia, Indonesia, and Philippines), tropical Pacific islands (Melanesia, Polynesia, and Micronesia) and westward to coastal India, Sri Lanka, East Africa, and tropical islands (e.g., Seychelles, Andaman, Mauritius) in the Indian Ocean. Many fibers are available in different parts of the coconut tree (Satyanarayana et al. 1982). The sheath is made up of an inner mat which is sandwiched between two layers of coarse fibers. Only preliminary studies of coconut leaf sheath fibers were reported in the literature (Satyanarayana et al. 1982). Though the fibers from many parts of the coconut trees are put to use, the sheath fibers are left as huge waste. In the present work, we separated the coarse fibers from the outer layers and the fine fibers from the inner mat to study their properties. The effect of alkali treatment on the properties of these fibers was studied using chemical, FTIR, WAXRD and TG analyses. Their tensile properties and morphology were also studied to assess their suitability as reinforcements.

Materials and methods

Extracted coconut tree leaf sheath fibers, sodium hydroxide pellets (Merk, India), benzene, sodium chlorite, acetic acid, sodium bisulphate and ethanol (S.d.fine-Chemicals, India) were used

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Extraction of the fibers from the trees

The Coconut Palm (*Cocos nucifera*) is a member of palm family (Arecaceae). Coconut leaf sheath fibers occur in mat form. The leaf sheaths collected from the trees were dipped in water for one week, thoroughly washed with tap water followed by distilled water, and dried in the sun for a week. Cleaned leaf sheath was separated to inner sheath mat and the outer layer fibers. The fibers of the inner mat and outer layers were separately kept in hot air oven for 24 h at 105−110ºC to remove the moisture. Some of these fibers were treated with 5% aqueous sodium hydroxide (NaOH) solution for one hour at room temperature, maintaining a liquor ratio of 25:1 to remove the hemicellulose and other greasy materials. These fibers were washed with water repeatedly and treated with dilute acetic acid to neutralise them. Finally the fibers were washed with distilled water before drying in hot air oven for a period of 24 h.

Morphology

The scanning electron micrographs of the surface of the fibers were recorded on a JEOL JSM 820 microscope. The micrographs of the cross section of the fibers were also recorded. These samples were gold coated before recording the micrographs.

FTIR spectral analysis

The two types of the fibers were cryogenically cooled and powdered separately. These powders were diluted to 1% using potassium bromide (KBr) and pellets were prepared. The FTIR spectra of the untreated and alkali treated samples were recorded in the 4,000–500 cm^{-1} region on a Perkin Elmer 16PC FTIR instrument with 32 scans in each case at a resolution of 4 cm^{-1}.

Chemical analysis

Chemical analysis of the untreated and alkali treated fibers was carried out as per the standard procedure (Chattopadhyay and Sarkar 1946; Sarkar et al. 1948; moran et al. 2008). In this analysis, the percents of α- cellulose, hemicellulose and lignin were determined. In each case, five samples were used and the average values were reported.

Thermo gravimetric analysis

The thermograms of the leaf sheath fibers were recorded on a Perkin Elmer TGA-7 instrument in nitrogen atmosphere at a heating rate of 10ºC/min in the temperature range of 50–500ºC.

X-ray analysis

The wide angle X-ray diffraction spectra of the leaf sheath fibers were recorded on a Rigaku Dmax 2500 diffractometer. The system had a rotating anode generator with a copper target and wide angle powder goniometer. The generator was operated at 40 KV and 150 mA. All the experiments were performed in the reflection mode at a scan speed of 4º/min in steps of 0.05º. All samples were scanned in 2 range of 5º to 50º.

Tensile properties

The tensile properties such as maximum stress, Young's modulus and % elongation at break of the leaf sheath fibers were determined using INSTRON 3369 Universal Testing Machine at a crosshead speed of 5 mm/min maintaining a gauge length of 50 mm. In each case, 10 samples were used and the average values reported.

Results and discussion

The photographs of the coconut tree with leaf sheath (marked with an arrow), leaf sheath, sheath inner mat and separated coarse fibers from the outer layers are presented in Fig. 1 a, b, c and d, respectively. The average dimensions of the outer layer (coarse) and inner mat (fine) fibers and the corresponding aspect ratio were presented in Table 1. From this table, it is evident that the coarse fibers had an average aspect ratio of 478 whereas fine fibers had a value of 3 322. Scanning electron micrograms of the surface and cross-section of the untreated and alkali fine and coarse fibers at different magnifications were shown in Fig. 2. From these micrographs it is evident that, for both types of fibers, surface of the fibers became rough on alkali treatment. Further, the micrographs of cross-section of the fibers indicate that the fibers had multicellular structure. Each unit cell of fibers was composed of small particles of cellulose surrounded and cemented together with lignin and hemicellulose. Similar observation was made in the case of some other ligno-cellulose fibers also (Rout et al. 2001; Mwaikambo and Ansell 2002; Ouajaj et al. 2004). Alkali treatment tends the fibers to react with the cementing material hemicellulose, and to increase the effective surface available for wetting by the resin when used in green composites.

Fig. 1 Photographs of (a) coconut tree with leaf sheath; (b) leaf sheath; (c) leaf sheath inner mat; (d) separated coarse fibers from outer layers.

Table 1. Average dimensions of the outer layer and inner mat fibers of coconut leaf sheath.

Fiber	Length (L) (cm)	Diameter (D) (cm)	L/D Ratio
Inner mat fine fiber	47.5	0.014	3322
Outer layer coarse fiber	47.5	0.099	478

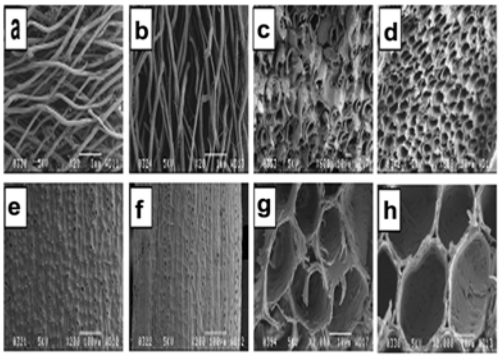

Fig. 2 Scanning electron micrographs of untreated and alkali treated coconut leaf sheath fibers. (a) Untreated leaf sheath inner mat fiber (20 X); (b) Alkali treated leaf sheath inner mat fiber (20 X); (c) Cross-section of untreated leaf sheath inner mat fiber (600 X); (d) Cross-section of alkali treated leaf sheath inner mat fiber (600 X); (e) Untreated leaf sheath coarse fiber from outer layer (200 X); (f) Alkali treated leaf sheath coarse fiber from outer layer (200 X); (g) Cross-section of untreated leaf sheath coarse fiber from outer layer (2000 X); (h) Cross-section of alkali treated leaf sheath coarse fiber from outer layer (2000 X).

The composition of both types of fibers was estimated by chemical analysis as per the method reported in recent literature (Moran et al. 2008). The chemical analysis of untreated and alkali treated fibers (Table 2) indicated the presence of α-cellulose, hemicellulose and lignin. Other compositions, usually regarded as surface impurities, were the pectin and wax. For comparison, the chemical compositions of some natural fibers are also presented in Table 2. From this table, it is also evident that the hemicellulose content of the fibers decreased on alkali treatment for all the fibers. Further, it can also be observed that chemical composition of coir sheath fibers was comparable with that of *Borassus* fruit fibers.

FTIR technique was employed to confirm the changes in the composition on alkali treatment of the coarse and fine fibers. The FTIR spectra of the untreated and alkali treated leaf sheath fibers are presented in Fig. 3. It can be observed that for alkali treated and untreated fibers, well defined bands corresponding to α-cellulose, hemicellulose and lignin were present in the spectra. But in the case of untreated leaf sheath fibers, additional bands at around 1 734 cm^{-1} and 1 248 cm^{-1} were also present corresponding to hemicellulose (Pandey 1999). On alkali treatment, these bands were found to be almost absent, indicating the elimination of the hemicellulose to larger extent. The bands at around 3 435 cm^{-1} and 2 930 cm^{-1} corresponded to α-cellulose whereas the remaining bands belonged to lignin. Thus, the FT-IR studies suggested the reduction of the hemicellulose content upon alkali treatment of the leaf sheath fibers. This was supported by the chemical analysis data of the alkali treated fibers as shown in Table 2. Almost similar trends were noticed for both the fine inner mat and coarse outer layer fibers.

Table 2. Chemical composition of some various natural fibers

Fiber	Chemical component					
	α - Cellulose	Hemicellulose	Lignin	α - Cellulose	Hemicellulose	Lignin
	Untreated			Alkali treated		
Ridge gourd						
a) Top layer	57.2	14	27.7	73	12.5	14.4
b) Bottom layer	60	14.1	25.9	72.7	12.3	14.96
(Rajulu et al. 2006)						
Tamarind	59	22	19	64.5	16	19.5
(Maheswari et al. 2008)						
Sterculia urens	62.9	24.3	12	81.5	7.5	10.8
(Jayaramudu et al. 2009)						
Polyalthia cerasoides	10.7	64.5	22.7	12.6	70.5	18.5
(Jayaramudu et al. 2009)						
Borassus						
a) Coarse	45.67	32.76	21.53	48.24	25.42	26.33
b) Fine	53.4	29.6	17	60.02	22	17.98
(Obi Reddy et al. 2009)						
*Coir sheath						
a) Inner mat	34.3	29.1	36.4	40.2	16.8	42.9
b) Coarse	53.6	22.3	24	60.5	12.5	26.9

The wide angle X-ray diffraction patterns of fine and coarse fibers are shown in Fig. 4. The diffractograms show two reflections, corresponding to 2θ values of around 16º and 22º, respectively. Among these, the low angle reflection (16º) was broad whereas the (22º) reflection was sharp and intense. These reflections were attributed to amorphous (I_{am}) and crystalline components (I_{002}) arising from hemicellulose and α-cellulose, respectively. The crystallinity index of the fibers was determined (Mwaikambo and Ansell. 2002) by using the following equation (1).

$$Ic = \frac{I_{(002)} - I_{(am)}}{I_{(002)}} * 100 \tag{1}$$

Where, $I_{(002)}$ (2θ=22º) represents the intensity of crystalline peak while $I_{(am)}$ (2θ=16º) denotes intensity of the amorphous peak in the diffractograms. Accordingly, the computed index values for untreated and alkali treated fine fibers from inner mat were 37.1% and 44.2%, respectively. These values for untreated and alkali treated coarse fibers were 39% and 41.5%, respectively. The increase in crystallinity of treated fibers might be due to loss of amorphous hemicelluloses as indicated in chemical and FTIR analyses.

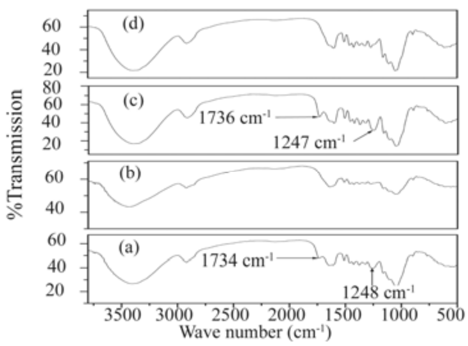

Fig. 3 FT-IR spectra of untreated and Alkali treated Coconut leaf sheath fibers. (a) Untreated leaf sheath inner mat fibers; (b) Alkali treated leaf sheath inner mat fibers; (c) Untreated leaf sheath coarse fibers from outer layer; (d) Alkali treated leaf sheath coarse fibers from outer layer.

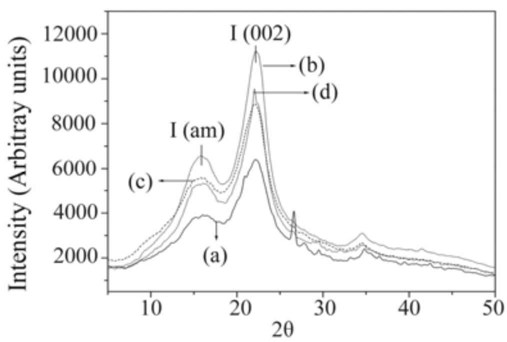

Fig. 4 X-Ray diffractograms of untreated and alkali treated coconut leaf sheath fibers. (a) Untreated leaf sheath inner mat fibers; (b) Alkali treated leaf sheath inner mat fibers; (c) Untreated leaf sheath coarse fibers from outer layer; (d) Alkali treated leaf sheath coarse fibers from outer layer.

The primary thermograms of both coarse and fine fibers before and after alkali treatment were presented in Fig. 5. Using these thermograms, the initial degradation temperature, 25% and 50% degradation temperatures, refractoriness (T*), inflection point (where the degradation rate was maximum) and integral procedural degradation temperature (IPDT) were calculated using the Doyle (1985) method. As shown in Table 3 in both cases, the initial and final degradation temperatures of the alkali treated fibers were slightly higher than those of the untreated fibers. However, for fine fibers, the IPDT and refractoriness of the fibers increased to a higher extent on alkali treatment. The inflection point was found to be unchanged for coarse fibers on alkali treatment. In the case of the fine inner mat fibers, the residue at 500ºC for untreated and alkali treated fibers were found to be 9.2% and 16.4%, respectively. Similarly, these values for coarse untreated and alkali treated fibers were 26.7% and 28.2%, respectively. Using the thermograms, the moisture content (%) was also calculated and these values are also presented in Table 3. For both coarse and fine fibers, the moisture contents were found to be slightly decreased on alkali treatment. The increase in thermal stability and decrease in moisture content of the fibers might be attributed to the increased crystallinity on alkali treatment as in crystalline polymers, the molecular are closely packed which lowers the permeation of water into them. Further, these results indicated that alkali treated fibers were suitable as reinforcement even in thermoplastic matrix materials with processing temperatures below 275ºC.

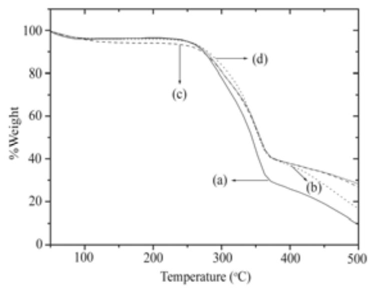

Fig. 5 Primary thermograms of untreated and alkali treated coconut leaf sheath fibers. (a) Untreated leaf sheath inner mat fibers; (b) Alkali treated leaf sheath inner mat fibers; (c) Untreated leaf sheath coarse fibers from outer layer; (d) Alkali treated leaf sheath coarse fiber from outer layer.

Table 3. Thermal degradation parameters of untreated and alkali treated coconut leaf sheath inner mat fibers and coarse fibers from outer layers

Degradation	sheath inner mat fibers		sheath separated coarse fibers	
Parameter	Untreated	Alkali treated	Untreated	Alkali treated
IDT	280.1	284.6	274.5	275
25%DT	305.7	323.2	313.8	314.6
50% DT	344.8	353.5	356.4	356
IP	352.6	342.1	352.7	352.3
IPDT	155.1	200	234	237.7
RF	184	266.6	371.5	3.9
MC	4.7	4.4	4.6	392.5

IDT: Initial degradation temperature; 25% DT: 25% degradation temperature; 50% DT: 50% degradation temperature; IP: Inflection point; IPDT: Integral procedure degradation temperature; RF: Refractoriness; MC: moisture content.

The tensile properties of the fine and coarse fibers are presented in Table 4. The data supported that for these fibers, the maximum stress, Young's moduli and %elongation at break increased on alkali treatment. As hemicellulose remained dispersed in the inter-fibrillar region separating the cellulose chains from one another for the untreated fibers, the cellulose chains were in a state of strain. When the hemicellulose was removed by the action of alkali, the internal strain was released and the fibrils became more capable of rearranging themselves in a more compact manner and resulted in a close packing of the fibers which led to improved tensile properties. The tensile properties of the coir sheath fibers are compared with those of some natural fibers in Table 4. From this table, it is evident that the tensile properties of coir sheath mat fibers were superior to those of some natural fibers.

Table 4. Tensile properties of some various natural fibers

Fiber	Tensile properties					
	Maximum stress (MPa)	Young's modulus (GPa)	Elongation (%) at break	Maximum stress (MPa)	Young' smodulus (GPa)	Elongation (%) at break
	Untreated			Alkali treated		
Hildegardia	46.4	2.3	2.84	57	3	3.3
Jagadeesh et al. 2008)						
Tamarind	61.16	2.1	6.22	66.26	3	7.97
(Maheswari et al. 2008)						
Sterculia urens	10.03	0.6	2	18.92	2	2.47
(Jayaramudu et al. 2009)						
Polyalthia cerasoides	44.3	3.4	2.5	51.6	2.7	5.7
(Jayaramudu et al. 2009)						
Borassus						
a) Coarse	50.9	1.2	41.2	53.5	1.6	41.9
b) Fine	65.2	4.9	47.2	90.7	9.8	58.5
(Obi Reddy et al. 2009)						
*Coir sheath						
a) Inner mat	119.8	18	5.5	128.6	6.8	8.7
b) Coarse	94.3	4.4	6.3	196.8	5.2	5.7

* Present work

Conclusions

The fine fibers from inner mat and the coarse fibers from outer layers of coconut leaf sheath were analyzed by SEM, FTIR, XRD and TGA techniques. The amorphous hemicellulose was found to be eliminated to large extent on alkali treatment. Due to this, the crystallinity of the alkali treated fibers was found to increase. Based on the thermal stability, the renewable and environment friendly natures, coconut leaf sheath fibers were found to be suitable materials as reinforcement in green composites.

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