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Millettia pinnata: a study on the extraction of fibers and reinforced composites

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Abstract

In this work, the potential for using *Millettia pinnata* stalk for extracting cellulosic natural fibers and its subsequent use in reinforced composites was studied. The extracted fibers were characterized for its composition, mechanical, thermal stability and morphological properties. Compositional analysis showed that the fibers possessed 54% cellulose, 12% hemicellulose, 15% lignin and 11% ash. The tensile strength of the fiber was 310 MPa, which is comparable to cotton and linen. The tensile strength of the *M. pinnata* fiber-reinforced polypropylene composites was 17.96 MPa which was similar to other natural fiber-based composites. *M. pinnata* fibers appear promising for a wide range of applications including textiles and other typical composites applications.

Keywords: Natural fibers, Biopolymers, Lignocellulosic material, Composites, Green materials

Introduction

Due to increasing awareness of the harmful effects of non-renewable materials, the world is gradually moving towards greener sustainable materials. On that line, natural fibers are being commonly used in various industries because of its high specific strength, low cost, low density, biodegradability and low-to-nil greenhouse emissions (Reddy and Yang 2009). Traditional fiber sources such as cotton, jute are resource intensive and are mainly grown for their fibers which limits the value added to the agricultural practice. Although these traditional sources dominate the world fiber market, a smooth supply of fibers cannot be ensured mainly due to the special growing conditions and extensive resources required. To overcome these limitations, other non-conventional sources of fibers should be considered for multi-fold benefits. One such source of non-conventional fibers is lignocellulosic agricultural residues and byproducts. Agro-residues are generated inevitably in huge quantities without any extra resources, but these valuable residues are generally burned leading to numerous environmental problems. Several researchers have studied the potential for using agro-residues and byproducts for various applications such as composites, water purification, bioproducts and so on (Guna et al. 2016, 2017, 2018, 2019a, b, c; Ilangovan et al. 2017, 2018a, 2019 Bhuvaneswari et al. 2017). Researches have also experimented the potential for using agro-residues for fiber extraction which seems to be a good technique to use these abundant raw material (Guna et al. 2019c; Ni et al. 2018; Kalita et al. 2019; Wang et al. 2016; Kale et al. 2018; Mohan et al. 2017; Vinayaka et al. 2017).

Millettia pinnata (*M. pinnata*) is a plant endemic to Asia and widely distributed in the Indian subcontinent. Locally it is known by various names such as 'Pongamia', 'Karanja', 'Honge' or 'Pungai'. *M. pinnata* seeds are widely used to produce biodiesel. Various parts of the tree are also used in traditional medicine and recent research has established its medicinal value including larvicidal activity (Srinivasan et al. 2003; Perumalsamy et al. 2015). However, the yield of essential oils and plant extracts are significantly low making it a non-viable application. For better value addition, alternate applications are to be found out. Using the plant as an alternate source of fiber or as reinforcement in composites is a straightforward and viable option.

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In this research *M. pinnata* fibers were isolated using the alkali solution method from the stalks. The fibers were characterized for its surface morphology, thermal stability, X-ray diffractometry, mechanical properties. The fiber and precursor compositional analysis was also carried out. Further, the extracted fibers were reinforced with polypropylene (PP) to fabricate composites and characterized for its mechanical properties. The properties of the fibers and the composites were compared to other lignocellulosic fibers and composites for potential textiles and composites applications.

Materials and methods

Materials

M. pinnata stalks were collected from S-Vyasa University Jigani campus. The stalks were washed with de-ionized water to remove any mud or dirt particles present, dried and used without any treatment. Laboratory grade sodium hydroxide and acetic acid for the extraction process were purchased from Sigma Aldrich, Bangalore. PP web was procured from Indian Oil Corporation, India.

Extraction of fibers

M. pinnata stalks were treated with 1 M NaOH solution for 90 min at 120 °C. The stalk-to-alkali ratio was maintained at 1:10. After treatment, the liquid containing dissolved hemicellulose, lignin and other extractives was decanted and disposed. The fibers extracted were thoroughly washed until the pH of the fibers was neutral. The neutral pH fibers were later immersed in 10% acetic acid solution for 10 min and then rinsed. The fibers were then dried at 110 °C for 3 h and stored for further characterization.

Fiber characterization

The cellulose and hemicellulose content of the stalk and the extracted fibers were carried out using the standard acid detergent and neutral detergent method as described in Guna et al. (2019d). Klason lignin was determined using the sulfuric acid hydrolysis method. Briefly, known weight of stalk and fibers were treated with 72% sulfuric acid (1:5 ratio of biomass:acid) for 2 h under constant stirring. The treated solution was then diluted using distilled water to 1:540 ratio and boiled under reflux for 6 h. After, the stipulated time, solution was filtered to obtain the residue. The obtained residue was washed and then dried at 105 °C for 6 h. The difference in weight was used to calculate the percentage lignin (Ilangovan et al. 2018b). ASTM E 1755-01 was used to estimate the ash content in the samples."

The fibers were tested for their tensile strength on a Universal Tensile Tester (MTS Mechatronics, Ichalkaranji, India) according to the ASTM D 3822-14 standard. Samples from three different sets of extraction were used for the characterization. 20 samples from each set, i.e., a total of 60 samples were tested. The mean and standard deviation was reported and compared with other standard cellulosic fibers.

Scanning electron micrographs of the *M. pinnata* stalk and fiber were obtained using a Hitachi SU 3500 scanning electron microscope at an operational voltage of 15 kV. Before observing, the samples were sputter coated with gold in an ion beam coater for 60 s.

The X-ray diffraction study was conducted using powdered samples of the stalk and fibers in a Bruker D8 Advanced Eco X-ray diffractometer equipped with Bragg–Brentano Focusing geometry. The analysis was done in a Cu-Kα radiation ($\lambda = 1.54$ Å) and the scattered radiations were recorded at 2θ angles varying from 10° to 65° using an SSD 160 detector. The readings were analyzed in Origin Lab software and the peak intensities were identified. The Segal method equation was used to calculate the crystallinity index as shown below:

$$X_c = \frac{(I_{002} - I_{am})}{I_{002}} \times 100\%,$$

where I_{002} and I_{am} are the peak intensities of the crystalline and amorphous materials, respectively (Segal et al. 1959).

A Shimadzu DTG 60 thermogravimetric analyzer was used to determine the thermal degradation of *M. pinnata* stalk and the extracted fibers. About 10 mg of the powdered specimen was placed in the analyzer and heated from room temperature to 600 °C at the rate of 20 °C/min. Air flowing at a rate of 50 ml/min was supplied during the analysis.

Fabrication of composites

Extracted fibers of *M. pinnata* were used as reinforcement in polypropylene composites. The fibers were carded using a carding machine several times to get a uniform mat of fiber. Sandwich type pre-pegs were prepared in 50/50 (w/w) ratio of fiber/PP and compression molded in a hot press at 170 °C for 120 s at 3000 PSI. Cold water was then let through the press and the samples were taken out once the temperature of the press dropped below 80 °C. The composite was then cut to ASTM D 638-14 standard for tensile tests and D 790-15 for flexural tests. The mechanical testing was carried out on a Universal Tensile Tester (MTS Mechatronics, Ichalkaranji, India) using appropriate load cells. At least 15 samples were tested for its tensile and flexural properties and the average and standard deviation was reported.

Results and discussion

Compositional analysis

Generally, composition of fibrous materials depends mainly on the function, growing and extraction conditions (Ilangovan et al. 2018b). Cellulosic fibers are covered by sheaths of lignin and hemicellulose. Upon treatment, the hemicellulose and lignin are washed away exposing the fiber bundles. The reduction in the percentage composition of hemicellulose is attributed to the cleavage of ester-linked substances of hemicellulose, particularly the α-(1–6) linkage in galactose which is sensitive to alkali treatment (Jeffries 1994). The alkali treatment of the *M. pinnata* stalk increased the percentage composition of cellulose in the fiber by 65% and decreased that of hemicellulose by nearly half its original value (Table 1). The lignin content was not significantly reduced as in the case of hemicellulose. It should be noted that excessive removal of hemicellulose will result in shorter fibers which may not be suitable for high-value applications.

Fiber tensile properties

During the extraction, fibers of lengths up to 6 cm could be extracted. The fibers were comparatively coarser than common textile fibers; however, they should be processable by regular textile-processing techniques. The tensile strength of the fiber was 310 MPa, slightly lower than that of cotton. The elongation of the fibers was found to be 1.83% which is comparable to linen. A comparison of mechanical properties of common cellulosic fibers and plant residue-based fibers is given in Table 2. *M. pinnata* fibers show promising potential to be used in textiles, composites.

Table 1 Composition of the *M. pinnata* stalks and fibers extracted from the plant

Composition	Stalk	Fiber
Cellulose, %	32.6 ± 1.6	54.6 ± 1.3
Hemicellulose, %	24.8 ± 1.0	12.3 ± 0.2
Lignin, %	18.3 ± 1.6	15.7 ± 1.3
Ash, %	16.6 ± 1.3	11.2 ± 0.6

Fiber morphology

The stalks had several impurities and deposits as can be seen from Fig. 1. Alkali treatment significantly removed the impurities; however, the impurities are not completely washed away. Further magnification of fibers shows smaller deposits on the fiber surface. Nevertheless, the surface is seen to be smoother compared to the unprocessed stalks. A more rigorous extraction method can lead to lesser deposits, but could potentially damage the fiber bundles. The fiber bundle width ranged from 50 to 70 μm with significant variations throughout the sample set. Individual fibrils ranged from 4 to 8 μm. The bundle width was comparable to the value reported from cotton stalk fibers (75 μm), rice and wheat straw (Reddy and Yang 2009). In the literature, individual fibrillar width of cotton ranged from 12 to 25 μm which was much higher than the *M. pinnata* fibers. The properties of any of the fibers are highly dependent on the length and width of the individual fibrils. Longer fibrils result in a finer fiber, whereas shorter ones lead to coarser fibers such as in this case. However, *M. pinnata* is not a conventional source of fiber and coarseness of extracted fibers is inevitable.

XRD analysis

The XRD plot of the stalk and *M. pinnata* fibers is given in Fig. 2. The diffractogram plot shows typical cellulosic peaks at the 2θ value 22.6°. These peaks are related to the crystalline structure of Cellulose I. The low diffraction intensity at a 2θ value of 18° characterizes the amorphous background. The percentage crystallinity of the stalk and the *M. pinnata* fibers were determined to be 53% and 57%, respectively. In comparison, cotton fibers had a percentage crystallinity of 62.3%. Also, the two smaller peaks at 14.9° and 16.6° seen in the cotton fibers were absent in the *M. pinnata* fibers attributed to the presence of hemicellulose and lignin particles in the latter. Such behavior was also observed in other non-traditional fibers (Reddy and Yang 2009). The crystallinity or ordered crystalline arrangements in cellulose fibers arises due to the intermolecular and intramolecular hydrogen bonding via hydroxyl groups. Here the crystallinity of the fiber increases as a result of the extraction process which could be attributed to the

Table 2 Comparison of the tensile properties of *M. pinnata* fibers and other common sources of plant fiber (Guna et al. 2019c; Ilangovan et al. 2018b)

Fiber properties	*M. pinnata*	Cotton	Linen	Switch grass	Jute
Tensile strength, MPa	310	392–511	667–885	715 ± 130	393–800
Tensile modulus, GPa	7.5	8.0–13.1	28.3–29.7	31.4 ± 9.5	10–30
Elongation, %	1.8	6.0–9.0	1.6–3.3	2.2 ± 0.7	1.5–1.8

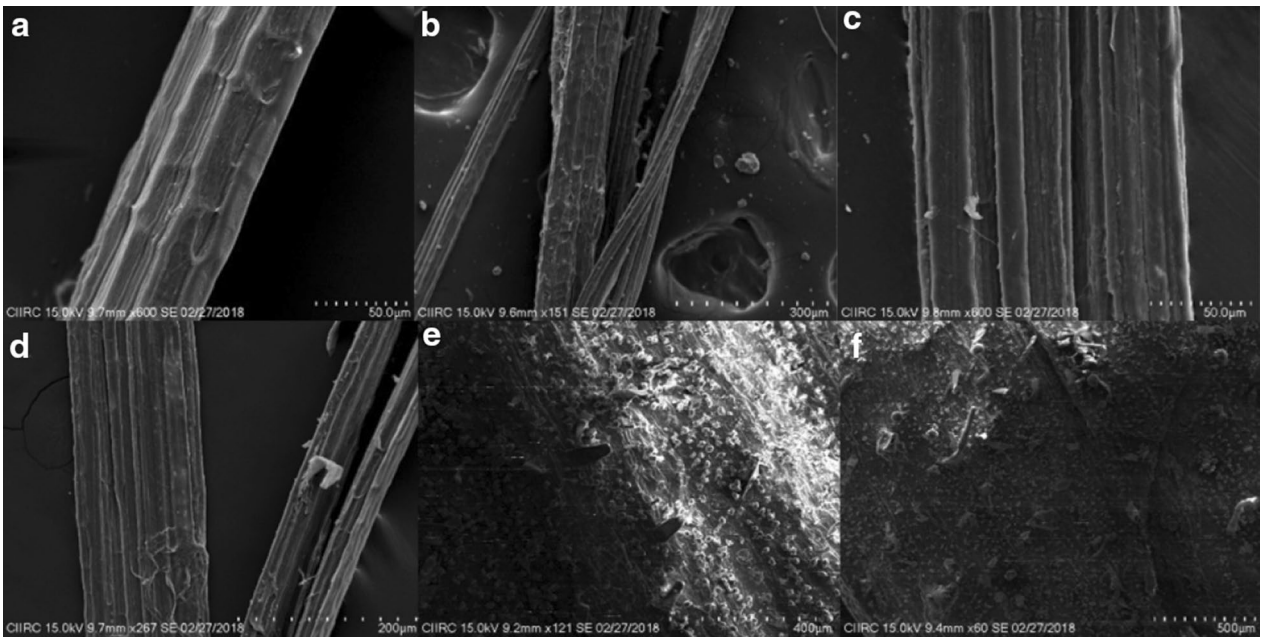

Fig. 1 Scanning electron microscope images of *M. pinnata* stalk and extracted fibers. **a**–**d** Surface features of the fibers, **e**–**f** surface features of the stalk

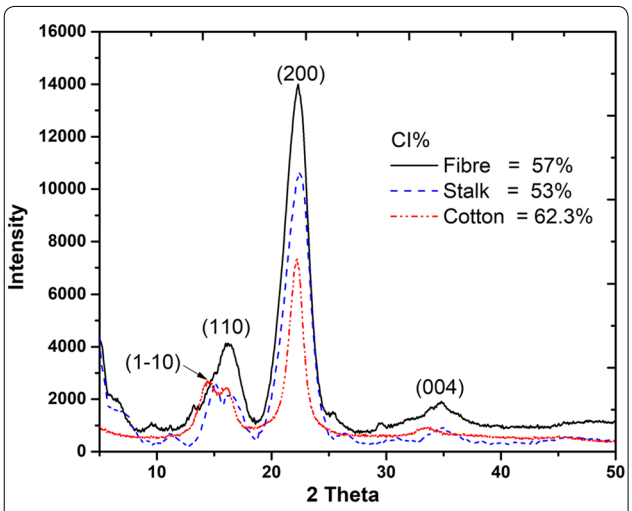

Fig. 2 X-ray diffractogram of *M. pinnata* stalk (before extraction) and extracted fibers compared with cotton fiber

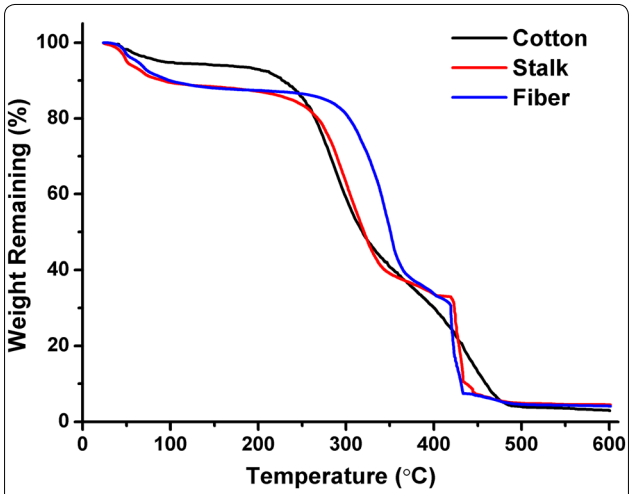

Fig. 3 TGA curves for the thermal degradation of *M. pinnata* stalks and extracted fibers compared with cotton fibers

removal of lignin and amorphous hemicellulose. Some additional peaks can be seen in the untreated stalk diffraction curve, presumably from minerals and impurities present in it. The value of crystallinity obtained in *M. pinnata* fibers compared to other non-traditional fibers is greater than that of turmeric plant residue (33%) and *M. pinnata* seed hull (47%), but less than that of Mengkuang leaf fibers (Kargarzadeh et al. 2012; Manjula et al. 2017; Ilangovan et al. 2018b).

Thermal stability

The results of the thermogravimetric analysis can be seen in Fig. 3. The untreated *M. pinnata* stalk starts degrading thermally at 260 °C, whereas the treated fibers start

Table 3 Tensile and flexural properties of 50/50 ratio of *M. pinnata*/PP composites compression molded at 170 °C compared with cotton stalk fiber and jute PP composites

Reinforcement	Tensile properties		Flexural properties		Reference
	Strength (MPa)	Modulus (MPa)	Strength (MPa)	Modulus (MPa)	
Cotton stalk fiber	15.7 ± 3.4	806 ± 140	12.4 ± 2.2	502 ± 99	Reddy and Yang (2009)
Jute	13.5 ± 1.3	979 ± 98	15.1 ± 1.0	620 ± 72	Reddy and Yang (2009)
M. pinnata	17.96 ± 1.1	875 ± 66	10.1 ± 0.6	580 ± 51	This work

showing thermal degradation only after 300 °C. The weight loss in the initial phase is attributed to water evaporation from the samples. The rapid thermal degradation seen after that from around 260–400 °C can be attributed to the thermal depolymerization of hemicellulose and pyrolysis of cellulose (Chen et al. 2011; Ludueña et al. 2011; Haafiz et al. 2013). The last streak of weight loss after 420 °C corresponds to the burning of ash. The final percentage residue recorded for the untreated stalks and the fibers were 7% and 2%, respectively. Similar residual percentage (3.8%) with the same three-step degradation mechanism was also observed in cotton fibers.

Composite properties

The mechanical properties of the composites developed are given in Table 3. *M. pinnata* fibers showed better tensile properties than cotton or jute-reinforced polypropylene composites, but lower flexural properties. The tensile performance could be due to the fact that the *M. pinnata* fibers are inherently stronger than cotton or jute. Since it is a coarser fiber, the area available for binding of polypropylene is considerably higher than that of cotton and jute. Although the properties of *M. pinnata* are compared to that of cotton and jute from other authors' works, the fabrication conditions are different. Hence, the comparison should be analyzed with caution. However, *M. pinnata* like any other natural fiber shows potential for use in composite applications and further work to determine the exact behavior of *M. pinnata* fibers will lead to a clearer understanding. Since, the morphological interactions are promising, we intend to carry out an extensive study of the mechanical properties of *M. pinnata* fiber-reinforced composites in another study.

Conclusion

Cellulose fibers were successfully extracted from *M. pinnata* plant using alkali treatment. Percentage cellulose increased after the treatment on stalks due to which the crystallinity of the fiber was higher than the stalk. The fibers had properties similar to that of other lignocellulosic fibers. There was an increase in the thermal stability of the fibers compared to the untreated stalk. Mechanical properties of the fiber-reinforced PP composites were comparable to other lignocellulosic composites. *M. pinnata* fibers can be potentially used in textiles and major composites applications. The use of extracted *M. pinnata* stalk fibers for other industrial applications can be further explored in future research considering the favorable properties of high crystallinity and thermal stability.

Abbreviations

M. pinnata: *Millettia pinnata*; PP: polypropylene.

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Authors' contributions

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Availability of data and materials

The datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

Ethics approval and consent to participate

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Consent for publication

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Competing interests

The authors declare that they have no competing interests.

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