International Journal of Bioresource Science
Citation: IJBS: **08**(02): 81-85, December 2021
DOI: 10.30954/2347-9655.02.2021.3

ASSOCIATION FOR AGRICULTURE ENVIRONMENT AND BIOTECHNOLOGY

Acidulated Extraction of Nanocellulose from Jute Fibre Wastes

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Received: 11-3-2021 **Revised:** 27-6-2021 **Accepted:** 24-11-2021

ABSTRACT

Cellulose is a unique organic molecule abundantly available in nature and can be a great source of useful synthetic nonmaterial for its potential applications as fillers in biodegradable nanocomposite used in automotives, packing and agriculture applications. In the present study, an attempt have been made to develop nanocellulose from the waste fibers that produced in the form of short length from jute spinning industry during several mechanical processes popularly known as "caddies". Sulphuric acid have been found be effective at a concentration of 60% for breakdown of cellulose long chain. Mechanical ultra-sonication in wet milling process yielded the optimum nanoform from alphacellulose. The extraction efficiency of alphacellulose from jute caddies is around 50% while the yield of nano cellulose is around 30%. The findings may mitigate problems of use of wood materials as a source of nanocellulose and find some alterantive source like jute stick and jute caddies. The process conversion to nanomaterials has wide implications in economic context and can minimize the long term industrial hazards through development of a valuable bio-resource.

HIGHLIGHTS

- An acidulated synthesis pathway of nanocellulose have been ventured through this study.
- Jute Caddies, a short fibre waste from jute industry was used as source for extraction.
- The extraction process from jute caddies to nanocellulose have been well documented.
- The derived nanocellulose have been characterized through particle size analyzer, surface morphology, FT-IR.

Keywords: Nonaocellulose, acidulated synthesis, Jute fibres, Caddies, nanocomposites

Natural fibres have relatively high aspect ratio, high strength to weight ratio, reasonable low density and good to moderate tensile strength and initial modulus (Abbasi and Baheti 2018). Cellulose is most abundantly available molecules in plant based natural fibres which contains both amorphous and crystalline structures. Several authors being enthusiast, has ventured for developing cellulose based high value products for industrial applications (Ghosh *et al*. 2018a, 2018b, Ray *et al*. 2017). Nanocrystalline cellulose (NCC), also called cellulose nanocrystals obtained from cellulose, is typically a rigid rod-shaped monocrystalline cellulose domain (whisker) with 1-100 nm in diameter and tens to hundreds of nanometers in length (Trache *et al.* 2020). NCC has a high degree of crystal structure (more than 70%), a very big length-diameter ratio (around 70), and a large surface area (150 m^2/g) (Tang *et al.* 2014). NCC can be used for many applications such as regenerative medicine, optical application, automotive application, composite materials and so on (Sharma *et al.* 2019). It can also be used in optical application (Revol *et al*. 1998), automotive application (Dahlke *et al.*

How to cite this article: Ray, D.P., Ghosh, R.K. and Das, I. (2021). Acidulated Extraction of Nanocellulose from Jute Fibre Wastes. *Int. J. Bioresource Sci.*, **08**(02): 81-85.

Source of Support: None; **Conflict of Interest:** None

1998), composite materials (Avella *et al.* 1993; Jiang & Hinrichsen, 1999) and so on.

In India, a huge quantity (around 4 Mt per annum) of jute (*Corchorus* spp.) stick is being produced as a primary by-product of jute fibre (economic part) cultivation (Nayak *et al*. 2013). Moreover, Indian Jute Industry generates nearly 40000 tonnes of processing wastes as by products commonly known as jute caddies (Nayak *et al*. 2012). In common practice these biomass are either burnt in boiler or used as such as firewood for domestic energy purposes. It is found from the experiment that if this stick/biomass of jute could be explored further as a new source of high-end value product, especially, nanocellulose, a huge return can be achieved. The incorporation of a small amount of nanometer-sized filler can yield composites with enhanced properties earnestly required for many industrial and technological applications (Whitesides, 2005). The conventional polymer-inorganic filler nanocomposites can have improved stiffness, strength, hardness and high temperature creep resistance compared to the unfilled polymers (Nguyen and Ishidah, 1987; Menendez and White 1984; Crivelli-Visconti, 1975)

In the present investigation an acidulation synthesis of nanosized cellulose was targeted which have wide industrial implications from a very cheap source of jute caddies, a waste materials from the jute Industry.

MATERIALS AND METHODS

Jute Caddies

Jute caddies were obtained from the jute mills and analysed separately for their components. The composition of jute caddies was determined by removal of oil and grease by extraction with trichloroethylene under reflux in a Soxhlet apparatus for 6 h, followed by opening and cleaning in a trash analyzer. The oil and trash (bark, jute stick remnants, etc.) free material was treated with 0.5 per cent hydrochloric acid to remove the remaining inorganic impurities.

Pre-treatment of Jute Caddies

The dried defatted caddies were successively undergone for delignification using sodium chlorite (1: 50 ratio) for 2 hr. The de-lignified materials (known as holocellulose) were the mercerized by 18% Sodium Hydroxide at room temperature for 120 min. The alkali-free dried samples (known as alphacellulose) were then taken for acid hydrolysis using 60% Sulfuric acid at 80°C for 60 min. The dispersion medium was deionised water. The acid hydrolysate was subjected to centrifuge for 20 min at 15000 rpm. The supernatant was discarded repeated to reach to the neutral pH. Finally the neutral suspension is sonicated in ice bath using Riviera Ultrasonicator for 15 min.

Fig. 1: Chemical modification of jute cadies to alpha cellulose

Characterization of nanocellulose

The particle size measurement of the milled material was carried out on Zetasizer nano series by Malvern® after the sonication. The dispersion medium was deionised water. The dispersion was ultrasonicated for 5 min before characterization whereas the refractive index was set at 1.52 to analyse the particle size. Single fiber strength of untreated jute fiber and chemically treated jute fiber was measured on vibroskop/ vibrodyn by Lenzing technik® with jaw speed of 10mm/min, gauge length of 10mm and tension weight of 500mg. Hence, in total 50 observations were recorded and mean value was calculated. Morphological study of the jute fibers after each phase of chemical treatment and ball milling was witnessed on scanning electron microscope (SEM) by TESCAN VEGA® at 30KV. Fourier Transform Infra Red (FTIR) analysis was carried out in Bruker Alpha-T model equipped with an attenuated total reflectance (ATR) device. The spectrum for each sample was recorded in the region of 500 – 4500 cm^{-1} at a resolution of 4 cm^{-1}

to confirm the removal of lignin after chemical treatment.

RESULTS AND DISCUSSION

The composition of jute caddies and jute sticks obtained from the commercial mill and from ICAR-NINFET were analysed and average values are presented in Table 1.

Table 1: Composition of jute caddies and jute sticks obtained from the commercial mill and from ICAR-NINFET

Constituents	Jute Caddies (%)	Jute stick (%)
Fat & wax	9.28	1.9
α-cellulose	61.05	40.80
Pentosan	19.61	22.10
Lignin	15.07	23.50
Ash	2.39	1.00

It is found that jute caddies contain high amount of waxy materials. This may be due to the treatment of jute with batching oils. The jute caddies contained 61.05% alpha cellulose whereas; jute stick contained nearly 41% alphacellulose. Jute stick contained grater amount of lignin (23.5%) than the jute caddies (15.07%). The pentosan content in both caddies and stick were more or less similar and ranged between 19.61 to 22.10%.

Acid catalysed hydrolysis of alpha cellulose

The dried alpha cellulose sample was subjected to acid treatment for further fractionation. Sulphuric acid 60% was found to be suitable for fractionation of alpha cellulose. The sample was treated in a magnetic stirrer for one hour. When there is a white precipitation in the reaction vessel the reaction is considered to be completed. It is then cooled and kept overnight. The precipitated sample is then subjected to rigorous centrifugation for 5-6 times at 15000 rpm till the pH of the reaction mixture becomes normal. At normal pH the reactant is taken to probe sonicator where it is treated for 2 hrs in interrupted sequences. The mixture becomes colloid and turned to be viscous mass which is subjected for DLS analysis.

Preparation of nanocellulose by neutralization of hydrolysate through repetitive centrifugation

The composition of jute caddies collected from different sources is presented in Table 2. It was observed that the caddies from NINFET mill yielded 57.49% alpha cellulose. Fractionation of this cellulose yielded 32.35% of cellulose in nano range. The commercial mill caddies were yielded 59.40% alphacellulose from which 30.24% nanocellulose obtained. In comparison the plain jute yielded 61.48% alpha cellulose which was converted to nanocellulose with a yield of 35.56 %.

Table 2: Composition of jute caddies collected from different sources

Caddies source	Initial weight (g)	Holo-cellulose yield (%)	α-Cellulose yield (%)	Nano-cellulose yield (%)
NINFET Mill	30	83.93	57.49	32.35
Commercial Mill	30	88.60	59.40	30.24
Plain Jute	30	88.25	61.48	35.56

Optimization of acid concentration efficient yield of Nanocellulose

The sulphuric acid concentration was optimized with correlation of yield of nanocellulose in specific nano range. Perusal of data presented in the chart showed that 60% concentration of sulphuric acid is optimum to yield the cellulose in the nano range. The higher concentration of acid did not show any significant results in terms of conversion of cellulose into nanocellulose. In higher concentration at 70% the samples were charred and no yield result could be obtained. The nano sized particle size was ascertained through dynamic light scattering.

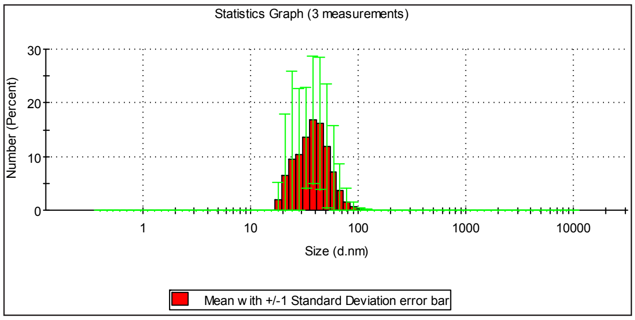

Fig. 2: Particle size determination of jute nanocellulose

Effect of Treatment on Mechanical properties of jute fibres

The Influence of treatment of chemicals on single fiber strength Mechanical properties of untreated jute fibers and after treatment with chemicals is shown in Table 3. Significant loss on modulus and tenacity was observed after chemical treatment. The reason was acid hydrolysis which was responsible for rupture of the bonds leading to deterioration in tensile strength considered to be useful in easier milling of the fibers.

Table 3: Mechanical property of jute fibres

Fibre type	Linear Density (dtex)	Youngs Modulus (g/den)	Tenacity (cN/Tex)	Elongation %
Untreated Jute	15.76	286.6	68.22	2.2
Treated Jute	15.27	56.25	52.36	5.7

Influence of treatment of chemicals on morphology

The morphological derivations in jute fibers after each chemical treatment are presented in Fig. 3. The jute fibers before treatment were bundles of individual strands held together by lignin but after mercerization and acid treatment, lignin was cleaned off partly and defibrillated the structure. After ultrasonication, a number of micro fibrils came out due to axial splitting of the fiber structure.

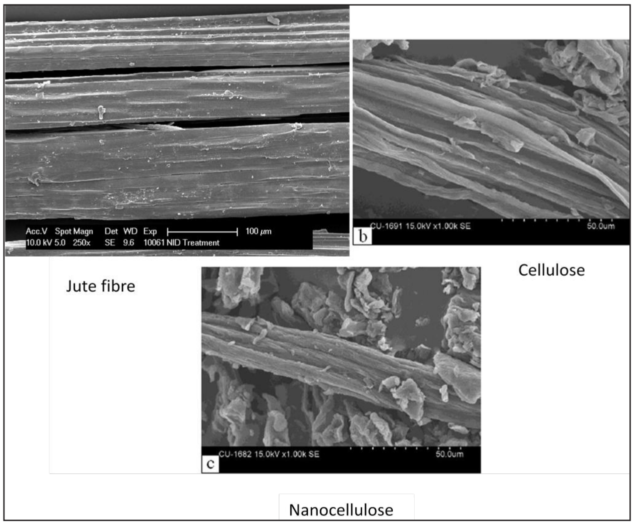

Fig. 3: Scanning Electron Microscopy of jute fibre, cellulose and nanocellulose

Influence of chemical treatment on structure of the fiber

The FTIR spectrums of untreated and chemically treated jute fibers are presented in Fig. 4. The typical peaks of lignin and cellulose lie at about 1000 cm^{-1} (due to C-O stretching vibration of the cellulose molecule) and 1500-1600 cm^{-1} (due to aromatic skeleton vibration of the lignin) respectively. It was observed that the characteristic peaks were situated in the same position after each treatment of chemical. However, the relative intensities of the peaks varied significantly. The ratio of 1500-1600 cm^{-1} to 1000 cm^{-1} increased, which confirmed that the content of cellulose increased and the content of lignin decreased.

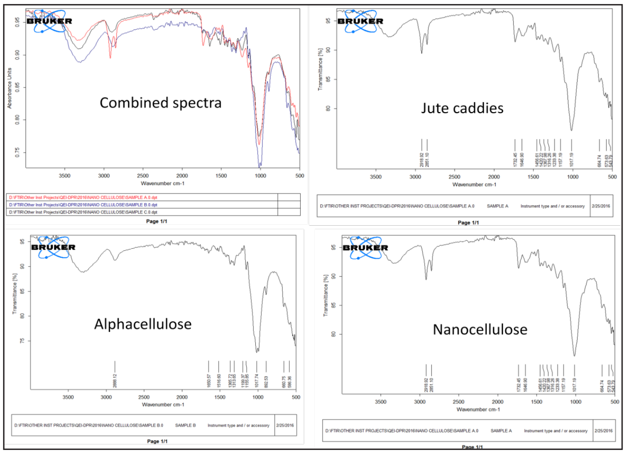

Fig. 4: FT-IR Spectra of different cellulosic forms obtained from Jute Cadies

CONCLUSION

Despite of the tremendous potentiality, jute fibre has some very traditional applications like preparation of hessian, sacking, carpet backing material and yarn (Ray & Ghosh, 2018). As the jute has high industrial value and availability is limited in the world, jute industry based other resource may generate a new avenue for alternate income. Chemical analysis of jute caddies and sticks reveals that jute caddies content more than 60% cellulose as compare to jute sticks. Moreover, the lignin content in jute stick is much higher and for this experiment, jute caddies were used for availability easy extractable celluloses. The sulphuric acid hydrolysis rendered to fragment the defibrillated celluloses into the nanoform and high speed sonication. The nanocellulose obtained from the experiment was further characterized for its particle size, determination, and surface morphology and FT-IR characterization. The yield of nanocellulose was found to be 30-40% on weight of dry cellulose. The method implores an economic conversion process to nanocelluloses which has wide implications in economic context and may further open a new horizon in vulnerable jute industry.

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