Indian Journal of Fibre & Textile Research
Vol. 45, September 2020, pp. 366-371

Novel chemical degumming process and its effect on structure and properties of mulberry fibres

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Received 16 July 2019; revised received and accepted 18 October 2019

A novel chemical degumming (Fibre-N) method has been used to extract fibres from the bark of mulberry plant. Fibres obtained by a new method (Fibre-N) are characterized by the X-ray diffraction, scanning electron microscopy, Fourier transform Infrared spectroscopy, residual gum content, fibre yield, and mechanical property measurement. Fibre-N method exhibits 17.71% lower residual gum content, 37.18% higher breaking strength, 18.36% higher breaking elongation and 10.81% higher fibre yield. Fibre-N process used to extract fibres from mulberry plant is more economical and efficient, which could be used as a promising method for producing textile yarns, nonwovens, and composites.

Keywords: Bast fibre, Chemical degumming, Mulberry fibre, Retting

Characteristic cellulosic fibres are delivered from different sources of plant, for example, bast, seed and leaf. The utilization of these fibres is extraordinary; for instance, apparel from cotton, jute for packing, fabrics and ropes from ramie fibres, etc. Paper mulberry is a tree in the family Moraceae[1]. The plant stalks of the paper mulberry are composed of bark and woody xylem[2]. The components of paper mulberry fibres are similar to cotton fibres, such as cellulose, hemicelluloses, lignin, and wax. Unlike cotton fibres, the paper mulberry fibres are composed of gum. The fibres were separated from the outer bark by boiling the bark in an alkali solution such as soda ash or caustic soda to dissolve the lignin, wax, and gums[3]. Fibres that are high in lignin are generally brown color. This is the characteristic of the paper mulberry fibres. Hence, the next step for preparation of the paper mulberry fibres is bleaching process. The purpose of this step is to whiten the fibres. The major bleaching agents used in preparation of the paper mulberry fibres are sodium hypochlorite or calcium hypochlorite[4]. Also, chlorine dioxide was used as bleaching agent by Lin *et al*[5]. The derivative of chlorine was claimed to be toxic chemical and related to the greenhouse effect[4]. To reduce the environmental impact, hydrogen peroxide is considered to be used instead of chlorine. The inner bark of the paper mulberry is composed of very strong fibres and can be used for making high quality paper. For using these fibres in textiles, the extraction of the fibres from the inner bark is very important. Also, the extracted fibres will be spun into yarns. The quality of the fibres has an effect on the mechanical properties of the produced yarns.

The present investigation has been aimed at introducing new chemical degumming method and applying them successfully in order to achieve high quality mulberry fibres to produce yarns by blending with cottons or without blending. Compared to flax yarns, mulberry fibre yarns have a tenacity 20% higher, an elongation 18% higher and an unevenness 30% lower. Cotton/mulberry fibre fabrics have softer and smoother hand than cotton/flax fabrics. This study also compared the properties of mulberry fibres obtained from Fibre-N with the retting process. The objective is to show the appropriateness of the Fibre-N process towards achieving a superior quality of fibres utilizing less amount of water and energy, which could make this process more economical and efficient.

Experimental

Materials

Raw materials (mulberry stems) were collected from Jute research institute, Bangladesh and then the leaves were removed. After collection, they were cleaned appropriately and stored under ambient condition. Chemicals used for this study, such as H_2SO_4 (98 % v/v), and H_2O_2 (30% v/v), were collected from Shanghai Macklin Biochemical Co. Ltd.

New Degumming (Fibre-N) Method.

Figure 1 represents the recipes and various stages of new degumming method.

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Retting (Fibre-R) Method

After collection, the stems were immersed in water for two weeks. The external skins of the stems were evacuated manually and finally mulberry fibres were separated. The extracted fibres were washed in running water and dried at 22°C for two days.

Fibre Properties

Fibres tensile properties were acquired utilizing an Instron fibre testing machine. ASTM method D2495 was utilized to evaluate moisture regain of the fibres. Standard test states of relative humidity and temperature (65%, 20°C) were maintained for every one of the tests. The density of fibres was calculated by pycnometer method using the following formula:

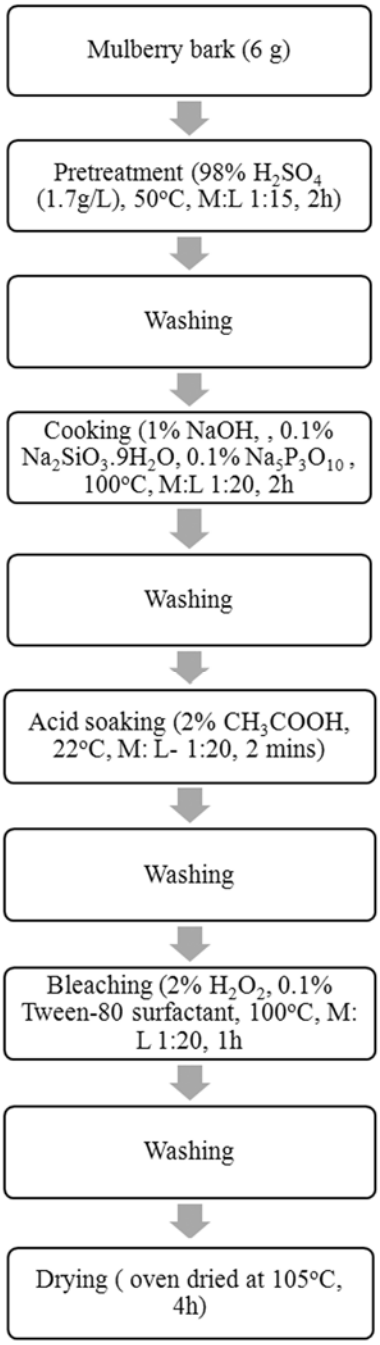

Fig. 1 — New degumming (Fibre-N) method

$$\rho_{mf} = \left(\frac{(m_1 - m_o)}{(m_2 - m_o)(m_3 - m_1)}\right)\rho_t \quad \ldots (1)$$

where m_0 is the mass of dry empty pycnometer in gram; m_1 , the mass of the pycnometer with fibres in gram; m_2 , the mass of the pycnometer with toluene liquid solution in gram; m_3 , the mass of the pycnometer with fibres and toluene solution in gram; ρ_t, the density of toluene liquid solution (0.867 g/cm^3); and ρ_{mf} , the density of mulberry fibres in g/cm^3.

Residual Gum Measurement

The method for investigation of residual gum content was done according to Das Gupta *et al.*[6]. Various steps followed in residual gum measurement method are shown in Fig. 2.

The formula for the determination of residual gum content (RGC) rate is shown below:

$$\%RGC = \frac{W_0 - (W_1 + W_2)}{W_0} \times 100 \quad \ldots (2)$$

where w_0 is the initial weight of the sample in grams; w_1, the final weight of the sample; and w_2 , the weight of dry filter mass after each step.

FTIR Spectroscopy

Infrared spectroscopy, as one of the basic strategies for molecular structure investigation, is generally utilized for the qualitative investigation of sub-atomic edifices. With the help of a Wiley mill, samples to be tested were made powdered and spectra were attained by utilizing Bruker Vertex 70 with a scan speed of 10 kHz, scan interval of 2 cm^{-1}, and scan range of 400-4000 cm^{-1}.

Physical Structure

ARL XTRA X-ray diffractometer was utilized to analyze the physical structure of the fibres by recording the diffraction patterns from $2\Theta = 3^o$ to 50^o. With the help of a Wiley mill, samples were powdered. Into a cavity holder, the powder was mounted so as to record the X-ray diffraction. The formula for the calculation of the degree of crystallinity as suggested earlier[7] is given below:

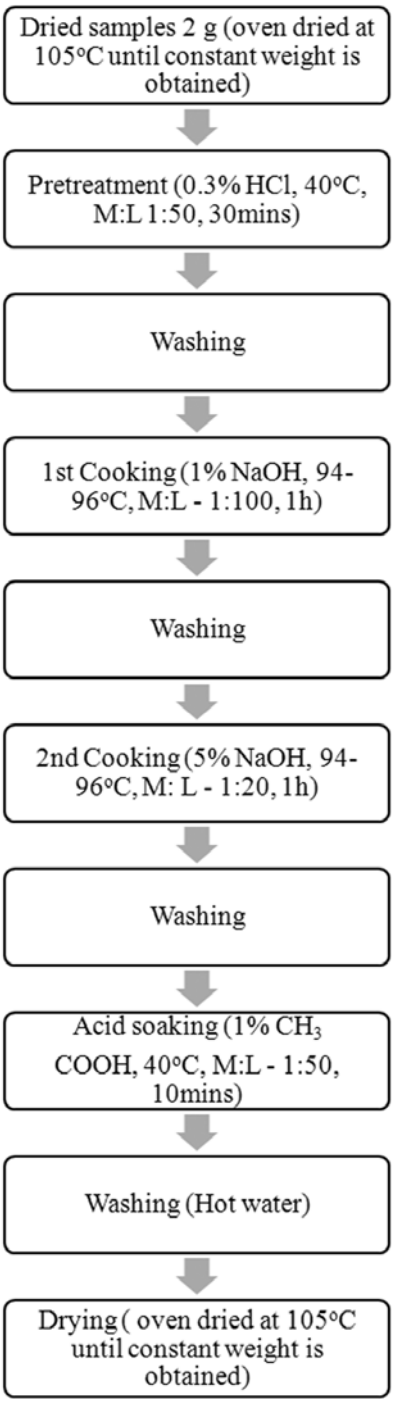

Fig. 2 — Residual gum content method

$$CI = (I_{002} - I_{am}) / I_{002} \times 100\% \quad \ldots (3)$$

where CI is the crystalline index; I_{002} is the maximum intensity of the (002) lattice diffraction; and I_{am}, the intensity diffraction at 18° 2Θ. The crystalline and amorphous materials in cellulose were indicated by the intensity readings at peak (002) and at 18° respectively[8,9]. Using the formula suggested by Hindeleh & Johnson[10] and Hindeleh[11], unit cell dimensions of cellulose crystal and the distances of lattice were determined. The formula for the calculation of crystallite size as reported by the Scherer[11] is given below:

$$D_{(hkl)} = k\lambda / B\cos\theta \quad \ldots (4)$$

where $D_{(hkl)}$ is the crystallite size; k, the Scherer's constant (0.89); λ , the wavelength of X-ray; and B, the full width at half maxima (FWHM) of (002) peak.

Morphological Structure

For the investigation of morphological properties of distinct fibre as well as fibre diameter, a Carl Zeiss SEM model v1tra55 was used. A Leica DM 2700M optical microscope was used to observe the diameter of fibres.

Results and Discussion

Fibre properties

Fibre tensile strength has an impact on yarn strength. Strength of yarn depends on two factors, viz fibre- to-fibre inner friction and fibre tenacity[12]. Commonly, natural fibres have a specific greater tenacity and lower elongation or the other way around. Fibres having higher modulus shows lesser extension when a given force is applied. Due to low modulus, cotton is more flexible and soft in comparison with jute and linen. Mulberry fibres look like cotton; however, are stronger than cotton[13,14]. Table 1 shows that mulberry fibres after Fibre-N process shows higher tenacity and elongation in contrast with Fibre-R and raw. This is because Fibre-N process is more efficient in removing cementing

Table 1 — Properties of mulberry fibres

Samples	Tensile strength cN/dtex	Elongation, %	Tensile modulus cN/dtex	Fibre diameter µm	Fibre density g/cm^3	Length cm	Residual gum content, %	Fibre yield %
Raw	4.33	4.69	90.85	26-46	1.09	2.6	-	-
Fibre-N	7.12	5.93	138.17	19-31	1.22	3.1	3.53	41
Fibre-R	5.19	5.01	119.31	23-39	1.14	2.8	4.29	37

matrix, which leads to fibre fibrillation and, in turn, increases the crystallinity of the fibres this leads to better mechanical properties. The densities of raw, Fibre-N and Fibre-R are 1.09, 1.22, and 1.14 g/cm^3 respectively. Density of fibre depends on the several factors such as environment, extraction process, soil condition, and plant age. For fabrication of light weight composites mulberry fibres can be utilized effectively as the density of fibre is less[14]. Variation in diameter can be observed from Table 1. It is evident that the diameter of Fibre-N process is smaller than those of Fibre-R and raw. This is because, diameter of fibre decreases due to removal of impurities from fibres.

Residual Gum Measurement

Properties similar to strength, flexibility, stretchiness, and water regain were determined by the chemical constituents, physical as well as the morphological structure of fibres [15]. In general, the amount of cellulose in cellulosic fibres obtained from nature lies in the range 60-95%. The extent of remaining constituents, such as lignin, pectin, hemicellulose, waxes, relies on the states of development, source of fibre, and the fibre extraction technique[15]. The amount of residual gum contents is tested to measure the optimal extent of the degumming process. The comparison of the fibre residual gum content on mulberry fibre produced by Fibre-N and Fibre-R process are shown in Table 1.

Generally, residual gum content and fibre yield rate are used to identify the effectiveness level of the degumming process. As shown in Table 1, the residual gum on fibre obtained by Fibre-N is found lower than Fibre-R. This is because, removal of pectin can be done completely by Fibre-R process but lignin cannot be completely eradicated. Even after the integration of acid pretreatment and peroxide bleaching with a high concentration of NaOH (Fibre-N), a specific quantity of residual lignin is always present in the fibres.

FTIR Spectroscopy Study

Figure 3 represents the FTIR spectra of mulberry fibres for Fibre-N, Fibre-R, and raw. From the spectra study, it is evident that the absorption peaks of 3419 cm^{-1} are attributed to OH group[16, 17]. Meanwhile, 2902 cm^{-1} and 2921 cm^{-1} absorption bands are attributed to CH and CH_2 stretching vibrations, that are clearly observed in Fibre-N and Fibre-R samples[17]. Furthermore, vibration peaks observed at

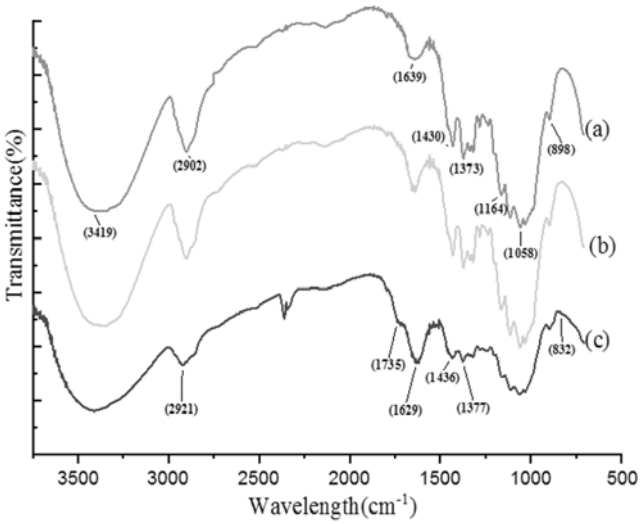

Fig. 3 — FTIR of mulberry fibres (a) Fibre-N, (b) Fibre- R, and (c) raw

1735 cm^{-1} in the spectrum of the raw, indicating that the C=O stretching of methyl ester and carboxylic acid or the acetyl group in hemicelluloses is not seen in the Fibre-N and Fibre-R spectra due to the successful removal of the pectin and hemicelluloses[17]. On the other hand, the absorption band at 1629 cm^{-1} of the raw is drastically decreased and shifted towards 1639 cm^{-1} for Fibre-N and Fibre-R, attributed to antisymmetric COO- stretching which proves the efficiency of degumming method to remove lignin from the raw fibres[16]. Moreover, shifting of absorption peaks is observed in the degummed fibres from 1436 cm^{-1}, 1377 cm^{-1} towards 1430 cm^{-1} and 1373 cm^{-1}, which denote CH_2 and CH symmetric bending mode respectively[17]. Besides, absorption peaks in the finger print regions at 1164 cm^{-1} and 1058 cm^{-1} are attributed to cellulose structure.[16, 17] In addition, absorption peaks at 1311 cm^{-1} and 898 cm^{-1} are associated with C-H scissoring and C-C bending[18, 19]. The small peak at 832 cm^{-1} , ascribed to aromatic C-H out-of-plane vibration in the lignin, is obviously decreased in intensity after the degumming process[20]. Consequently, it may be due to the reason that the absorption peaks of all the non-cellulose substances, for instance, hemicellulose, pectin, lignin, and wax-like substances, are totally missing or radically decreased for Fibre-N than that of Fibre-R and raw fibres.

Physical Structure

Fibres physical structure depends on the quantity of amorphous and crystalline substance, their alignment

to the fibre axis and crystallite size. Different properties are observed in celluloses like cotton, ramie, and wood, although they have similar structures of polymer and unit cell[21]. The reasons for having dissimilar properties can be due to the distinctions in the direction of the crystalline and amorphous sections regarding the fibre axis, crystallite size, quantity of crystalline and amorphous substances, and presence and category of non-cellulosic material[21].

In order to get a clear idea about the crystal structure of mulberry fibres, the samples have been tested by XRD, and their patterns are studied (Fig. 4). It is evident that raw sample has cellulose I structure[22] having diffraction peaks at 15.2°, 17.2°, 21.2°, 22.96°, and 34.5°, which are allocated to (101), (10$\bar{1}$), (021), (002) and (040) planes individually. This proves that the crystal structure of mulberry fibre has not been altered by the application of different methods. Compared with raw, the diffraction peaks strength of (101), (10$\bar{1}$), (002) and (040) are found sharper in the samples Fibre-N and Fibre-R, representing that the treated mulberry fibres have a superior crystal structure. It indicates that the proportion of the amorphous region in fibres decreases due to the removal of fibre impurities during degumming. Crystallinity index (CI) for the raw sample is 55.07%. Nevertheless, the CIs are 76.63 and 69.14% for Fibre-N and Fibre-R respectively, which are less than that of flax (80%) but higher than that of cotton (60%) [23]. The results show that the crystallinity is in the order: Fibre-N> Fibre-R> Raw Fibre. This marvel can be clarified by the way that the amorphous area of cellulose in strands is dissolved amid their treatment with NaOH while their crystalline structure is adjusted and the crystallinity of cellulose raises.

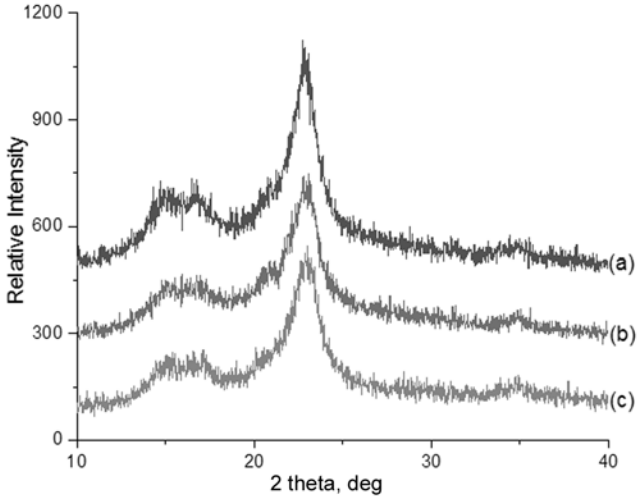

Fig. 4 — Diffraction intensities of (a) Fibre-N, (b) Raw, and (c) Fibre-R

Morphological Structure

Similar to other bast fibres, mulberry fibres are bounded by the protective layer made up of hemicellulose, lignin, pectin, etc [Fig. 5(a)], which helps to save it from being harmed by the external environment. Figure 5(b) depicts the fibre bundles obtained by Fibre-N method. Figure 5(c) shows individual fibres of *A. venetum* extracted from the Fibre-N process with an even and spotless surface since the majority of the impurities are evacuated, and its length is also well enough to be utilized for textile applications. Figure 5(d) indicates the surface of Fibre-R that has a rough layer of outer surface with little impurities. Nevertheless, the outer layer of the fibre containing impurities is well evacuated, and the surface of the Fibre-N becomes smoother than that of Fibre-R due to the removal of majority of non-cellulosic components, which would result in improvement of spinnability.

In this study, efforts have been made to supplant the Fibre-R technique by Fibre-N. Effects of chemical degumming on the surface chemistry, molecular structure, morphology, and physical properties of mulberry bast fibres are studied. XRD, FTIR, and SEM demonstrate the most noteworthy chemical changes on the surfaces for Fibre-N. SEM photomicrographs illustrate that Fibre-N processes are successful in removing waxes and unwanted surface impurities. XRD shows gradual changes in crystallinity for different samples. After chemical treatment, the crystallinity of Fibre-N increases, which results in more tensile strength, higher fibre yield, and less residual gum content in contrast with fibres obtained by Fibre-R. The experimental results show that fibres obtained from Fibre-N have the characteristic cellulose I arrangement and properties superior from Fibre-R process, which makes it more suitable for direct textile and other applications. Moreover, Fibre-R process is harmful to both fibres and environment as the waste water contains microorganisms. Hence, Fibre-N method is more efficient, fibre-friendly and could be used as a promising method for bulk production of mulberry fibres for manufacturing spun yarn, nonwovens, and composites.

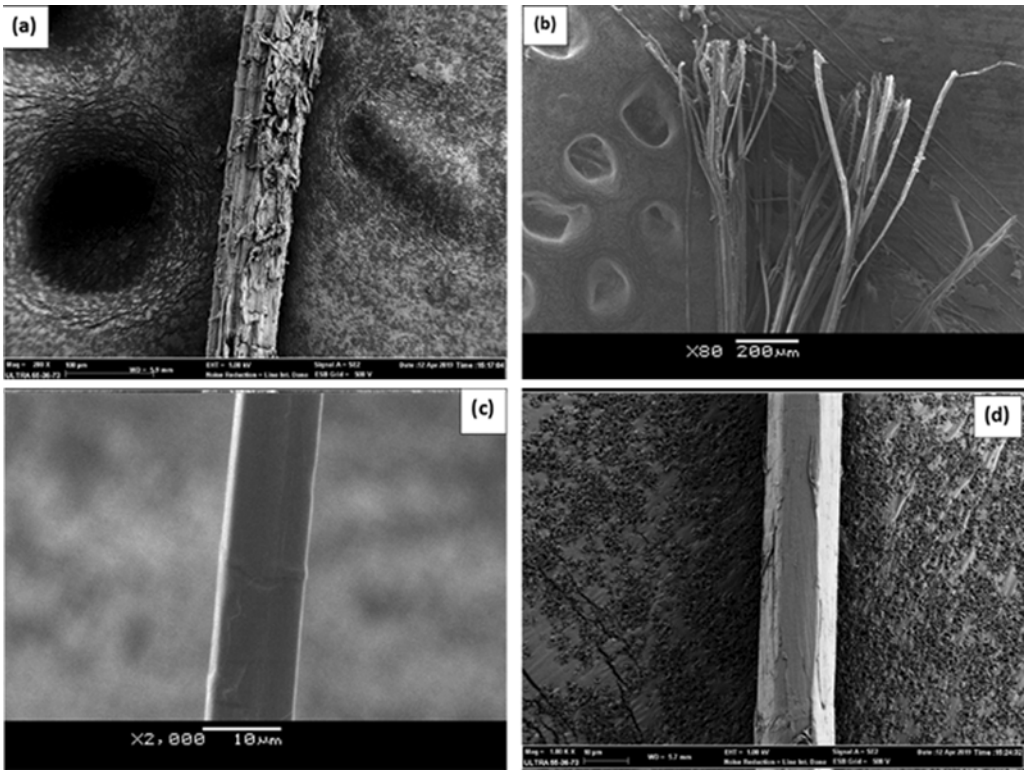

Fig. 5 — SEM micrographs of mulberry fibres (a) raw, (b) Fibre-N bundle fibres, (c) Fibre-N single fibre and (d) Fibre-R single fibre

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