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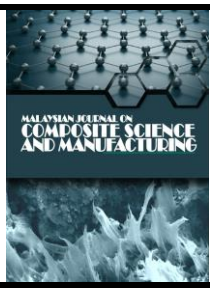

Characterization of Chemomechanical Treated New Fibrillated Cellulosic Nano Fiber from Spent Tea Leaf

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Qumrul Ahsan[1,*], Wo Angeline[2,3], Lin Wei Qi[2], Zaleha Mustafa[1], Noraiham Mohamad[1]

[1] Smart Material Research Group, COSSID, Fakulti Kejuruteraan Pembuatan, Universiti Teknikal Malaysia, 76100 Melaka, Malaysia.
[2] Fakulti Kejuruteraan Pembuatan, Universiti Teknikal Malaysia, 76100 Melaka, Malaysia
[3] Friedrich-Alexander-Universität, Erlangen-Nürnberg, Martensstr. 5a, 91058 Erlangen, Germany

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ABSTRACT

This project focuses on the fibrillation of cellulose nano fiber (CNF) derived from spent tea leave (STL) and the characterization of the extracted fibers. The new extraction technique was aiming to realize the synergistic effects of chemical treatment and mechanical fibrillation process parameters (blending speed and time) for yielding of CNF from STL. STL underwent chemical (NaOH and HCL) pretreatment to degrade the intermolecular bonding between cellulose, hemicellulose and lignin followed by fibrillation at various blending speed and time to isolate the CNF contents mechanically. Vacuum filtering was utilized to filter the fibrillated fibers. The fibrillation effects on the extracted fibers were characterized and analysed by using FTIR, optical and scanning electron microscope. The results indicated that the average diameter of fibrillated fibers were decreased with the concurrent increase of blending speed and time. It was found that blending time and speed in mechanical fibrillation have substantial effect on yielding of nanofibers. Fine STL fibers blended at speed and time of 23500 rpm and 10 min provide the smallest CNF diameter (within the range of 20 to 40 nm) along with better fibrillated yield of 15.22% than coarse STL fiber.

Keywords:
Nano Fibrillation; Spent Tea Leaf;
Blending Speed; Cellulose, Morphology

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1. Introduction

Natural plant fibers are the main resources of the agricultural lignocellulosic biomass comprised of about 10-25% lignin, 20-30% hemicellulose, and 40-50% cellulose which provides major mechanical strength in plant cell walls due to its stiffener like structure [1]. Most cellulosic materials consist of crystalline and amorphous domains in varying proportions based on the sources of cellulosic biomass. Recently the use of natural plant fiber in its nanocellulosic form has shown

* *Corresponding author.*
E-mail address: qumrul@utem.edu.my

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growing attention because of its lightweight, cost effective and ecological advantage as renewable.

Nanocelluloses are extracted either from wood or plant fiber principally through chemical hydrolysis, mechanical disruption, enzymatic, or combination of two (or more) approaches [2-4]. Ideally, nanocellulose can be obtained from any plant fibres rich in cellulose e.g. oil palm empty fruit bunches, bamboo pulps, jute fibers, kenaf and others or extracted from woods [5]. Chirayil et al. [6] reported that NFCs can be extracted from algae, tunicates and bacterial cellulose but the main source is natural plant cell walls. In general, nanocellulose could be categorised into two groups, one with longer semi-crystalline fibrillar structure called cellulose nanofibers (CNF, or nanofibrillated cellulose), while the other one with highly crystalline structure in the name of cellulose nanocrystal (CNC, or nanocrystalline cellulose). CNFs are cellulose chains rich in plant cell wall, having a complex intramolecular hydrogen-bonding crystalline structure.

Usually, CNF was most often obtained through a top-down hydrolysis approach in which lignocellulose, lignin and hemicellulose were removed on a layer-to-layer basis. Most of the reactants penetrate only the amorphous regions on the surface of the crystallites with a low level of arrangement order and leaving the intracrystalline regions unaffected. Anwar et al. [7] emphasized that pretreatment is a crucial step for the recovery of cellulose content from lignocellulosic biomass. The purpose of the pretreatment is to enhance the accessibility of the cellulose. Therefore, in order to increase cellulose accessibility, the pretreatment of cellulose fibers has to be carried out to destroy the physical structure of the biomass by destroying the lignin barrier, destroying the crystallinity of the cellulose, and removing the non-cellulosic components. In order to improve the reactivity of the cellulose, hemicellulose and lignin are required to decomposed [8], therefore chemical pretreatment methods have been developed to remove hemicellulose and lignin and reduce the crystallinity of cellulose to enhance the biodegradability of cellulose. Cellulose, hemicellulose and lignin from lignocellulosic can be degraded by various chemicals such as alkali, acids, salts, oxidants and solvent. Basically, the most common used chemical pretreatment are acid and alkali based hydrolysis [7]. As reported by Damisa [9], the combination of acids and alkali are more effective in pretreatment of lignocellulosic waste than acid and alkali alone. Acid treatment also known as delignification, or bleaching process, which use to remove most of the lignin and other impurities on the surface of the fiber. Phanthong et al. [10] pointed out that the solid product obtained after the acid treatment is defined as holocellulose, mainly including hemicellulose and cellulose in the fiber. However, alkali treatment is used to remove the amorphous polymer of hemicellulose and the remaining of lignin after acid treatment. The fiber product obtained from alkali treatment is predominantly in the form of cellulose and other non-cellulosic materials have been removed. Alemdar & Sain [11] found that the cellulose content in wheat straw was increased effectively from 43% to 84% by applied alkali and acid treatment on wheat straw.

Mechanical nanofibrillation is the another approach to breakdown cellulosic fibers into nanofibers. Uetani & Yano [12] have obtained cellulose microfibril bundles as individualized cellulose nanofibers with diameters of 15 nm to 20 nm by mechanical fibrillation. Conventional methods of mechanical fibrillation employing devices such as grinding, high-pressure homogenizer and microfluidizer which are closed systems and impossible to observe the disintegration of fibers. Hence, it is necessary to adopt a method with an open system that enables the observation of fibrillation mechanism and also fibrillates pulps as effectively as previously mentioned conventional methods. Uetani & Yano [12] used wood pulp to disintegrate into uniform CNF using a high-speed blender and the degree of fibrillation were compared with the CNF produced by a grinder. They found that the pulp treated for 30 min in the blender showed the same degree of fibrillation with less damage to the CNF compared with the pulp treated in the grinder. The optimum pulp concentration and agitation speed during fibrillation in blender method is found to be 0.7 wt.% of pulp at 37000 rpm.

Chaker et al. [13] experimentally investigated the comparison of delignified cellulose fibers from alfa and sunflowers with different hemicellulose contents to prepare CNF by mechanical disintegration using a high-pressure homogenizer and a domestic blender. They found that as the hemicellulose content increased, fibrillation is also increased. Besides, the fibers from sunflower containing thinner cell walls give rise to a higher yield in fibrillation. Researchers [14] recommended mechanical process combined with chemical pretreatment for reducing energy in fibrillation process and they found that jute fibers could be defibrillated under the shearing action of ball mill friction and then refined into nanofiber segments under the impact force of mechanical blender.

In terms of fiber morphology, Uetani & Yano [12] found in the FE-SEM image that the pulp treated in blender for 30 minutes showed the same degree of fibrillation with less damage to the CNF compared to the pulp treated in the grinder. It also showed that the pulp fraction was reduced and the fibrillated portion increases as the agitation time increases. Davoudpour et al. [15] claimed that morphology in FE-SEM image can be varied with the physical properties of fibers by changing its isolation process, processing parameters, and cellulosic fiber source. Mohanty et al. [16] studied the changes of fiber molecular structures before and after the chemical treatments in FTIR spectroscopy. For the chemically treated fiber, the disappearance of the C=O stretching frequency corresponding to the carbonyl group can be attributed as the removal of lignin from the fiber surface.

Tea leaf fiber is a unique natural fiber with fresh aroma and rich in phenolic extractive content which contribute to be more resistant against fungal and termites [17] and is considered as a new resource for research because of its certain properties such as low density, high specific stiffness, renewability and biodegradability. Bajpai *et al*. [18] stated that tea leaf is mainly composed of 37wt% of cellulose, hemicellulose and 14 wt.% of lignin. Spent tea leave (STL) is a waste product in tea leave production process and that waste is extracted after drying, chopping and roasting of leaves. A large amount of STL would be left over every day and could be easily collected with very low cost. However, the biggest challenge in fibrillation process is to maximum yield of cellulose without damaging the nanofibrils. Therefore, it is essential to adapt new chemomechanical treatment by which fibers are partially or fully fibrillated.

This paper is an effort to study the viability of extracting nanocellulose fiber from spent tea leave (STL) by chemical pre-treatment and fibrillation method, and the characterization of the extracted STL fiber. In chemical treatment, the sodium hydroxide solution (NaOH) and hydrochloric acid solution (HCL) were used to weaken the intermolecular bonding between lignin, hemicellulose and cellulose of industrial tea leaves followed by fibrillation to isolate the lignocellulosic contents in mechanical blender with varying blending speed and time to obtain higher yields of nanocellulose from STL. Afterwards centrifugation method was utilized to determine the yield of nanofibrillated cellulose (CNF) which was characterized and analysed by optical and scanning electron microscope to study the morphology of fibers.

2. Methodology

2.1 Spent tea leave (STL)

BOH Plantations Sdn. Bhd. located at Cameron Highland, Pahang, Malaysia provides the STL for this research. The STL provided is the residues during tea production process, which mainly consists of the stalks and fibers of the tea plant. Fibers collected from stem and branch of tea plant are termed as coarse STL (CSTL) whereas fibers derived from tea leaves are designated as fine STL (FSTL). Both types of fibers were characterized by microscopic observations as well as chemical analysis.

2.2 Chemical Pre-treatment and Pulverisation

Course and fine STL fibers of 5.0 grams respectively were soaked into 500ml of 6 wt.% sodium hydroxide solution (NaOH) for 3 hours at room temperature, in order to raise the surface area of cellulosic fibers and become more susceptible for hydrolysis. Afterwards, the respective grades of STL fibers were immersed into 500ml of 3.7 wt.% hydrochloric acid solution (HCl) at 70°C for 2 hours to solubilize the hemicelluloses. Again, the respective hydrolysed fibers were immersed into 500ml of 2 wt.% NaOH solution for 2 hours at 70 °C to disrupt the lignin structure and breakdown the pectin. In between each chemical alteration, the fibers were washed thoroughly with running water and checked the fibers neutrality between pH 6.5 to 7.3 by a pH meter. Finally, STL fibers were dried and were pulverized in a rotor mill (Pulverisette 14, Germany) with rotational speed of 7 rpm. The dried-blended STL were granulated with 3-micron meter sieve.

2.3 Fibrillation and Vacuum Filtering

Herein, the structural components of the pretreated STL fibers were broken down using a Vita-Mix blender, model 5200 with a maximum blade speed of 23500 rpm. First, 285 millilitres of water were added into 2 grams of fibers making a suspension with fiber contents of 0.7 wt.% in a 2 litres jar size blender. The suspension was blended at a low blade speed setting of 1000 rpm to obtain even fiber distribution and then agitated at three different high speed setting of 5200 rpm, 13500 rpm and 23500 rpm for 1 minute, 5 minutes and 10 minutes. All STL fiber samples were vacuum-filtered using a polytetrafluoroethylene membrane filter (0.1 μm mesh) through the porcelain Buchner funnel.

2.4 Yield in fibrillated cellulose

The yield of fibrillated cellulose was obtained from centrifugation method by a centrifuge machine where the suspension was centrifuged at 3800 rpm for 30 minutes to separate the fibrillated material from the non-fibrillated or partially fibrillated ones that settled down as sediments. Later the sediment fraction was collected by vacuum filtering and was dried in a drying oven for 24 hours at temperature of 40°C. The yield percentage of fibrillation was calculated by using the Equation 1 [13]:

$$Yield\ \% = \frac{Dry\ Fiber\ Weight - Dry\ Sediment}{Dry\ Fiber\ Weight}\ x\ 100 \quad (1)$$

2.5 Characterizations of STL Fiber

2.5.1 Optical Microscopy (OM)

The appearance of as received and fibrillated coarse and fine STL fibers were observed by optical macroscope at magnification of 5. In addition, the length and diameter of the chemically treated and fibrillated STL fiber were measured by optical microscope, model Zeiss Axioskop 2 at magnification of 50.

2.5.2 Scanning Electron Microscopy (SEM)

A SEM (A Zeiss SEM type Evo 50 Series) operating at accelerating voltage of 5 kV and magnification in the range of 50 -1000 was used to investigate the fiber surface morphology before and after chemical treatment of the coarse and fine STL. All samples were coated with gold by

Quorum mini sputter coater before SEM observation.

2.5.3 Field Emission Scanning Electron Microscopy (FESEM)

To obtain a higher resolution of the fibrillated cellulose which is up to 1 nm, field emission scanning electron microscope of model Joel JSM-7600F was used. Low accelerating voltage and magnification of range of 12 – 100,000X was set to get the high resolution image of the fibrillated cellulose.

2.5.4 Fourier Transform Infrared Spectroscopy (FTIR)

The changes of functional groups of STL before and after chemical treatment and fibrillation were analyzed by FTIR. The analysis was carried out by JASCO FT/IR-6100 that can perform up to 20 scans per second with maximum resolution of 0.5 cm^{-1} and the functional groups of composite were analyzed within the spectra range from 4000 to 400 cm^{-1}.

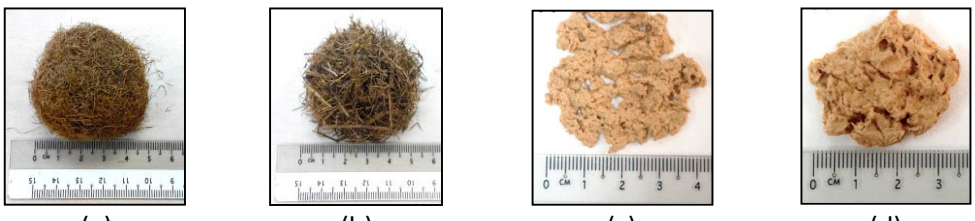
3. Results

3.1 Characterisations of STL Fibers by Optical Microscopy

The length and diameters of two different grades of STL fiber are shown in Table 1. Among two, it was found that FSTL is the finest grade fiber as it possesses highest aspect ratio with lowest diameter and length. The macroscopic appearances (Figure 1) of fine/coarse STL fibers after fibrillation clearly show the lumpy conditions as compared to received ones which indicate that the downsizing of fiber diameters and lengths lead to agglomeration.

Table 1
Physical properties of STL fiber

STL Fiber Grade	Average Diameter, µm%	Average Length, µm	Aspect Ratio
FSTL	130	7,330	57
CSTL	650	10,450	16

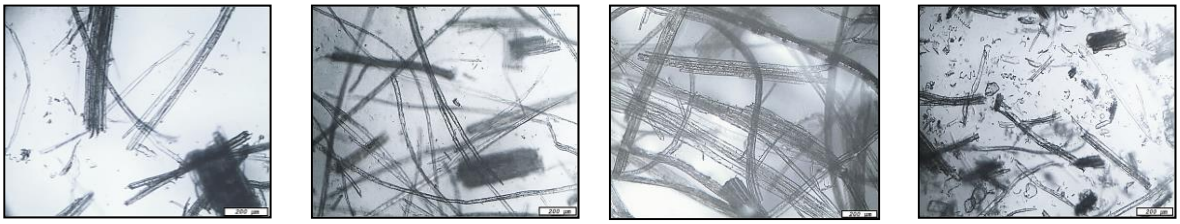


(a) (b) (c) (d)

Fig. 1. Macro photographs of (a) as-received FSTL, (b) as-received CSTL, (c) fibrillated FSTL and (d) fibrillated CSTL fibers

The average value and standard deviation of fibers diameter of as-received, pretreated and fibrillated STL were investigated qualitatively and quantitatively by optical microscope The OM images of coarse and fine fibers were compared at different blending times with the lowest blending speed (5200 rpm) are presented in Figure 2. The fine STL has comparatively higher aspect ratio [Figure 2(a)] and the more technical fibers associated with the destruction of fiber bundles to microfibrils were separated out with the increment of blending time [Figure 2(b)]. Whereas the coarse STL fibers

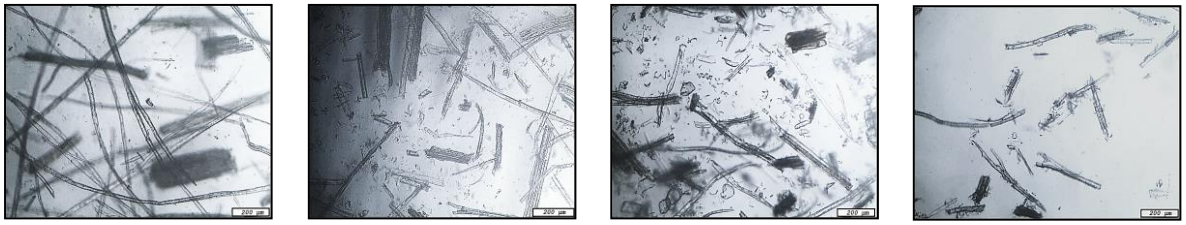
were separated out easily for blending time of 1 min [Figure 2(c)] but broke down into smaller length during fibrillation as the blending time increased [Figure 2(d)]. This may attribute to the effective chemical pretreatment on fine STL fibers to remove more hemicellulose and lignin contents. Thus, the fine STL fiber separation ability is increased [16].



(a) (b) (c) (d)

Fig. 2. Fibrillated STL fibers blended at speed of 5200 rpm with blending time of (a) 1 min and (b) 10 min for fine and (c) 1 min and (d) 10 10 min for coarse fiber

Figure 3 illustrates the effect of blending speed on both coarse and fine STL by fixing the blending time of 10 min. Figure 3 (a) and (b) show that the fine STL are susceptible to have better fibrillation and higher aspect ratio with increment of blending speed. The coarse STL initially formed low length fiber [Fig. 3(c)] but higher blending speed may result a significant agitation effect that could isolate the technical cellulose fiber from coarse STL [Fig. 3(d)]. In addition, higher blending speed may generate a significant agitation effect that can isolate the technical fibers from STL.

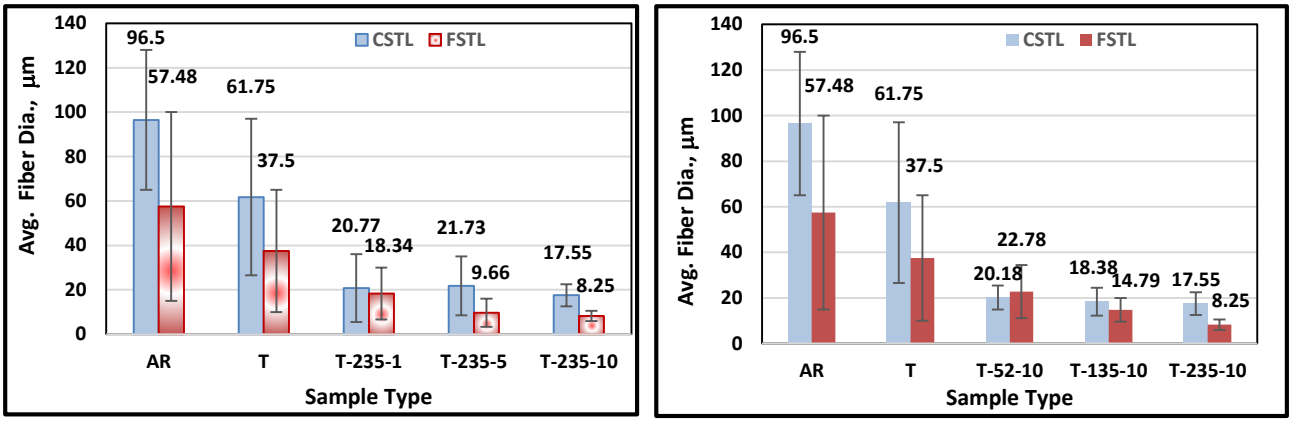


(a) (b) (c) (d)

Fig. 3. Fibrillated STL fibers blended at time of 10 minutes with blending speed of (a) 5200 rpm and (b) 23500 rpm for fine and (c) 5200 rpm and (d) 23500 rpm for coarse fiber

At least 50 fiber diameters of the coarse and fine STL were measured from optical micrographs using quantitative metallographic software. The average diameter and standard deviations of the fibers after different blending time and speed were calculated and plotted as shown in Figure 4 (a) and (b) respectively.

Referring to Figure 4, the average diameter of the as-received STL coarse and fine fibers were reduced by 36.0 % from 96.5 µm to 61.75 and 34.7 % from 57.48 µm to 37.50 µm respectively after the acid-alkali treatment. During HCl treatment, most of the lignin and other components are removed from the surface of STL fiber, whereas, most of the amorphous hemicellulose and the remaining of lignin was hydrolysed and become water soluble during the high temperature NaOH treatment [10] and thus reduction in STL fiber diameter occurred. Figure 4 also shows the effect of blending time or speed on the average diameter of coarse and fine STL. By maintaining the maximum blending speed and time of 23500 rpm and 10 min, the maximum reductions of fiber diameter are 88.8 % and 85.6 % for coarse and fine STL fibers respectively. It infers that whatever the condition of as received STL fibre either as coarse or fine, diameter reduction could be carried out efficiently with increase of blending speed and time.



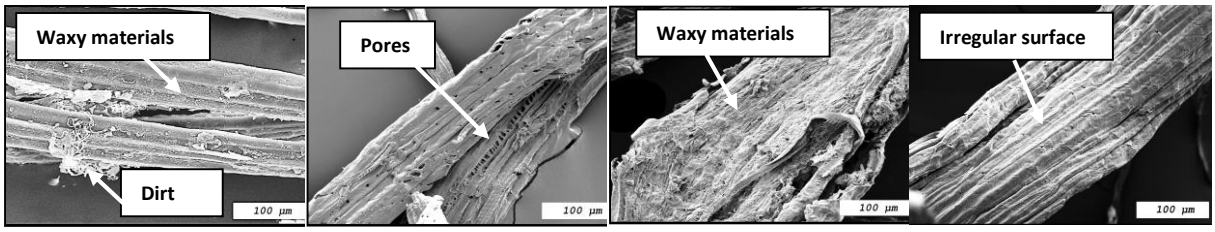
(a) (b)

**AR- As received, T-Treated, 52/135/235 are rpm in x100 and 1, 5 and 10 are minutes*

Fig. 4. Average values of fiber diameters of coarse and fine fibrillated STL at (a) 23500 rpm with different blending time of 1 min, 5 min and 10 min and (b) 10 min with different blending speed of 5200, 13500 and 23500 rpm

3.2 Characterisations of STL Fibers by SEM

The surface morphologies of as-received and chemically treated coarse and fine STL fibers characterized by SEM are shown in Figure 5 (a) to (d) respectively. From figures, the as-received coarse and fine STL fiber bundle is covering with dirt and waxy substances. After the chemical pretreatment, the surfaces of both coarse and fine STL fiber bundle are cleaned and dirt-free having wavy in shape because of the irregular arrangement of the microfibrils [19]. Due to the removal of hemicellulose and lignin, both coarse and fine STL fiber bundle increased in porosity with some holes and pores between the bundles. Adhesive layer of the coarse STL has been removed by leaving a porous skeleton-like structure of hemicellulose connecting the technical fibers.



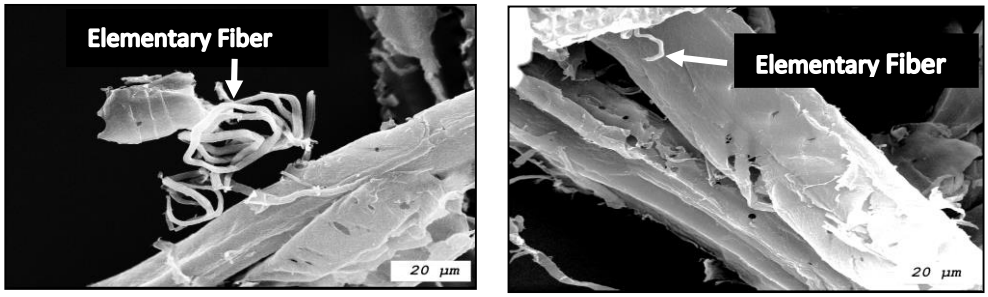
(a) (b) (c) (d)

Fig. 5. The SEM micrograph of (a) as-received coarse STL (b) chemically treated coarse STL (c) as-received fine STL and (b) chemically treated fine STL fibers

The effect of fibrillation towards the extraction and yielding of technical fibers by blending the coarse and fine STL at the maximum speed and time (23500 rpm and 10 min) were also micrographed (Figure 6) by SEM. Figures clearly show the fibrillation of coarse and fine STL fibers. The elementary fiber, termed as single fiber in which a number of threads like microfibrils are bound together separated from the technical fibers and more microfibrils were coming out from elementary fibers as the blending time increased. This may due to the effective removal of lignin and hemicellulose of the STL during the chemical pretreatment.

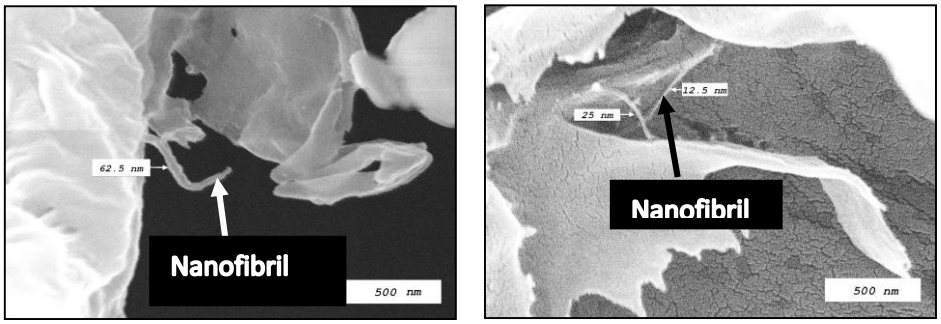
In order to observe further, the presence of nanofibrils or cellulose nano fiber (CNF) in both coarse and fine STL, the Field Emission Scanning Electron Microscopy (FESEM) was used to analyse the fibers blended at maximum blending speed of 23500 rpm and maximum blending time of 10 min.

Figure 7 indicates the presence of nanofibrils or CNF in coarse and fine STL and the presence of nanofibrils or CNF derived from natural plant fibers those have typically a diameter of about 10 nm to 30 nm. CNF fibers of such diameters with made up of 30–100 cellulose molecules were also reported by Mohanty et al. [16]. Comparatively, the fine STL fibrillated nanofibrils have smaller diameter. It is suspected that due to the physical structure of fine STL, removal of hemicellulose and lignin are more susceptible for the elementary fibers that fibrillated into mesofibrils and later nanofibrils.



(a) (b)

Fig. 6. SEM of fibrillated elementary fiber of (a) CSTL and (b) FSTL at a blending speed of 23500 rpm and blending time of 10 min



(a) (b)

Fig. 7. FESEM of fibrillated nanofibril of (a) CSTL and (b) FSTL at blending speed of 23500 rpm and blending time of 10 min

3.3 Yield of Nanofibril or Cellulose Nano Fiber(CNF)

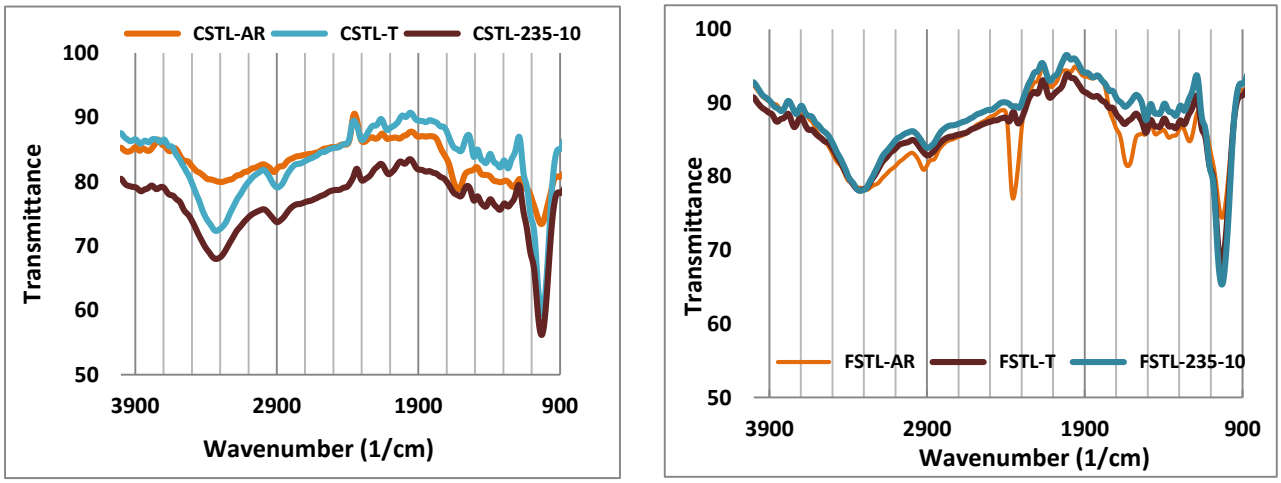
In order to evaluate the extent of fibrillation, the yield of fibrillation fraction was determined by centrifugation method. This approach was shown to give a good indication about the degree of fibrillation of the cellulose nano fibers [13]. Equation 1 in section 2.4 calculated the fibrillation yield percentage and the average value of three replications are shown in Table 2. Result shows that the fine STL has a higher average yield compared to the coarse STL which relates to the smaller diameter of fibrillated nanofibrils as observed in FESEM. Alternatively removing greater proportion of hemicellulose by chemical pretreatment will bring the microfibrils into a closer contact, which favours their strong interaction through hydrogen bonding [13]. Thus, it renders that the fibers were difficult to fibrillate and required longer time for fibrillation.

Table 2
Average yield percentage of nano fibril from coarse and fine STL

Nomenclature	Yield (%)	Average Yield (%)
CSTL-T-235-10	15.70	14.68 ± 1.00
	13.71	
	14.63	
FSTL-T-235-10	18.85	15.22 ± 4.17
	16.13	
	10.67	

3.4 Fourier Transform Infrared Spectroscopy (FTIR)

Figure 8 (a) and (b) show the comparisons of FTIR spectrums of as-received/treated/ fibrillated fibers for coarse and fine STL respectively. In figures, the band at 3031 cm^{-1} (BM-AR) and 3002 cm^{-1} (SW-AR) is due to aliphatic and aromatic C-H stretching which is a characteristic functional group in any organic molecules like the cellulose, hemicellulose and the lignin existing in natural fibers [20] In Figure 8(a) the fibrillated coarse STL show a sharper and more intense of aliphatic and aromatic C-H stretching at band 3031 cm^{-1}, whereas the fine STL in Figure 8(b) does not show any significance change in spectrum for that band when compared with as received fibers. Camposo et al. [20] stated that this situation may due to greater activity of some C-H bindings which may infer that the pretreated coarse STL fiber may retain more hemicellulose composition and lead to more amorphous structure as compared to pretreated fine STL. Meanwhile, the band at ~2900 to 2950 cm^{-1} represents C-H stretching of methyl or methylene groups on the fibers [21].



(a) (b)

Fig. 8. Comparison of FTIR spectrums of as-received, chemically pre-treated and fibrillated (23500 rpm, 10 min) for (a) coarse and (b) fine STL fiber

Bands near 1370 cm^{-1} to 1330 cm^{-1} are assigned as the in-plane C-H bending, may derive from hemicelluloses or cellulose in as received fiber [4]. The near disappearance of these bands for both pretreated coarse and fine STL fibers may be attributed as the removal of the hemicelluloses. However, the bands from 1330 cm^{-1} to 1270 cm^{-1} of fine STL which representing the main components in lignin are flattening and the peak is the smallest. Hence, it can be concluded that for pretreated, the fine STL have a better removal of lignin as compared to coarse STL. In addition, the lower absorbance in chemically pretreated of coarse and fine STL means that the lignin was removed

and the cellulose content would be higher than that in the as-received fiber. Finally, the spectra resulted from the functional groups of the coarse and fine STL have no significant changes after the fibrillation process. This indicates that the no changes of functional groups occur during the chemical pretreatment process instead of fibrillation process which was also reported by Ahsan et al. [22].

4. Conclusions

The chemical pre-treatment and mechanical fibrillation by varying blending speed and time were carried out to extract cellulose nano fibers from coarse and fine STL followed by characterisation of extracted fibers. OM clearly revealed that the fibrillated coarse STL fibers tend to break down into shorter length as the blending time increased during fibrillation. Whereas the fine STL fibers retain its length longer along with high aspect ratio. SEM morphology reveals that chemical pretreatment helps to remove hemicellulose and lignin from both STL fibers. After fibrillation, presence of elementary fibers in coarse STL isolated individually, whereas the fine STL fibers fibrillated into even smaller mesofibrils as the blending time increased. However, FESEM micrographs revealed that the nanofibers from fine STL are within the range of 20 to 40 nm in diameter representing better fibrillation as compared to coarse STL fiber. This finding is also supported by the higher fibrillation yield of fine STL fibers obtained from centrifugation method. From the changes of functional groups and bond stretching of the fibers obtained from FTIR spectrum, pretreated fine STL is found to have lower bond activity of hemicellulose and lignin, which may lead to high crystallinity. Hence, it can be concluded that the fine STL comparatively is easier to be fibrillated than the coarse STL.

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