Nanofibrillated Cellulose from Banana Pseudostem Reinforced Bagasse Paper Sheets

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Abstract. Underutilized agro-waste of cultivated banana pseudostem was treated by a soda pulping process and passed through a microfluidizer to prepare nanofibrillated cellulose (BA). TEM images confirmed the presence of thin fibrils and fine webs of BA with average diameter and length of 20.0±7.6 nm and 1.7±0.1 µm, respectively. Reinforcing potential of this BA nanofibers in recycled papers was investigated by adding 0.5, 1.0 and 5.0 wt% BA into the recycled bagasse sheets. Tensile testing results indicated that all mechanical properties (i.e. tensile index, breaking length, Young’s modulus, and elongation) were noticeably increased all at once. SEM images revealed that the physically retained BA nanofibers in the sheet structures bridged and linked between bagasse fibers increasing inter-fiber bonding, hence, strengthening the sheets as a result. Toughness of the reinforced sheets was also found to be significantly enhanced (~128% increase with 5.0 wt% BA incorporation). The change in failure mode showing fibers stretching and slippage before breaking was observed on fractured surface of this paper sheet. This BA cellulose nanofiber demonstrated to be highly promising as reinforcement in pulp and paper industry.

1. Introduction

The recovery of paper and board is a well-established practice these days as a result of concerns about environmental issues, energy saving, population growth, and the shortage of wood supplies [1-2]. By increasing the number of recycling cycles, however, fibers experience structural damages due to mechanical refining and hornification phenomena which reduces paper quality, especially its strength [3]. Recently, an addition of cellulose nanofibers has emerged as a new approach to improve the strength of paper and board due to their attractive characters such as high specific surface, high aspect ratio, high intrinsic mechanical properties, renewable nature, biodegradability, high potential availability, and capacity to be functionalized [1]. The incorporation of cellulose nanofibers has been reported to significantly increase bond degree and paper strength as well as extend the life span of paper products to more than twice [3]. The main methods currently used to effectively produce cellulose nanofibers or nanofibrillated cellulose are the high pressure homogenization and the micofluidization [4]. Cellulose nanomaterials are usually extracted from wood, however, fibers from annual crops and agricultural residues has attracted more and more interest due to their wide abundance, low cost, and availability [5].

The main goal of this study is to examine the potential of nanofibrillated cellulose prepared from banana pseudostem (BA) to be used as a reinforcing material in recycled bagasse papers. Mechanical
and surface properties as well as porosity of the reinforced paper sheets, which are interesting from an industrial point of view, were characterized.

2. Experimental

2.1. Materials

Cultivated banana pseudostem was taken from Mae Fah Luang University Botanical Garden, Thailand. Bleached bagasse sheet was supplied by Biodegradable Packaging for Environment Public Co., Ltd., Thailand. Sodium hydroxide (NaOH) was an AR grade from QRë C, New Zealand.

2.2. Preparation of nanofibrillated cellulose from banana pseudostem (BA)

Firstly, the banana pseudostem pulp was prepared by soda pulping process. The pseudostem was chopped into 10 cm length then washed and dried in a hot air oven until constant weight. Next, the dried pseudostem was size-reduced by using a pulverizer (Nimut Engineering Co., Thailand) and treated with 18% NaOH at 98±2 °C for 30 min using the liquid:solid ratio of 10:1. The obtained pulp was then rinsed by tap water to remove black liquor until cleaned and pH adjusted to 7. The pulp was disintegrated by using a kitchen blender (House Worth model HW-BDC2PC) at 15,000 rpm for 1 min before screening by the metal meshes no. 18 and 200 with opening sieve sizes of 1 mm and 74 µm, respectively. The collected pulp for further use was the screened portion between the two metal meshes. To prepare nanofibrillated cellulose, the previous pulp was diluted to 0.5 wt% and homogenized through the interaction chamber of 87 µm at 25,000 psi for 20 passes using an M-110P microfluidizer® processor (Microfluidics, U.S.A.).

2.3. Preparation of bagasse pulp

The bagasse sheet was torn into small pieces (ca. 1 cm × 1 cm) and dispersed in tap water at 2% consistency overnight. Then, the bagasse suspension was disintegrated by using a kitchen blender (House Worth model HW-BDC2PC) at 15,000 rpm for 5 min before screening by the metal meshes no. 18 and 200. The screened pulp portion between the two metal meshes was collected and prepared into a suspension with 2% consistency for further use.

2.4. Preparation of bagasse and bagasse/BA paper sheets

The bagasse pulp slurry was directly added to the BA suspension in ratios of 0.1, 0.5, 1.0, and 5.0 wt% and gently stirred and mixed for 1 min. Then, each mixture was partially dewatered by hand press between the two metal meshes no. 400 and hot-pressed into a paper sheet at 105 °C for 5 min under 159 kPa pressure. The bagasse sheet without BA addition was also formed for comparison. The grammage of all paper sheets made were in the range of 220-280 g/m² having density and porosity in the ranges of 0.81-1.00 g/cm³ and 34.4-47.1 %, respectively.

2.5. Bagasse pulp and BA characterization

The bagasse pulp fibers were characterized by scanning electron microscopy (SEM) (LEO 1450 VP, U.S.A.) at an accelerating voltage of 10 kV. Gold was sputtered on samples prior to the observation. The morphological analysis of the BA nanofibers was carried out using a transmission electron microscope (TEM) (Hitachi HT7700, Japan) at an accelerating voltage of 80 kV. For sample preparation, a BA suspension of 0.001 wt% was deposited to a copper grid with formvar film and the droplet was maintained on the grid for 5 min, and then strained with a 2% aqueous uranyl acetate. The diameter and length of bagasse pulp and BA fibers were measured from SEM and TEM images, respectively. The reported values were averaged from 50 bagasse fibers and 10 BA fibers.

2.6. Paper sheet testing and characterization

Tensile testing of the paper sheets was carried out using Instron (model 5566) testing machine (U.S.A.) equipped with a 1 kN load cell. The width and gauge length of specimens were 10 mm and 30 mm, respectively. The cross-head speed was set at 15 mm/min and the test started with a preloaded of 1 N. Before testing, all specimens were conditioned at 50% relative humidity (RH) at 25 °C overnight.
The surfaces and fractured surfaces (after the tensile test) of the paper sheets were characterized by using SEM (LEO 1450 VP, U.S.A.) at an accelerating voltage of 10 kV. A thin layer of gold was coated on sheet samples prior to the examination.

3. Results and Discussion

Figure 1 shows SEM (a) and TEM (b) images of bagasse pulp fibers and BA nanofibrillated cellulose fibers, respectively. The bagasse pulp fibers had an average diameter of about 11.7±4.9 µm and length of 274.8±105.9 µm with the aspect ratio around 29. On the other hand, after passing through the microfluidizer (20 passes), the BA nanofibers with an average diameter of 20.0±7.6 nm and length of 1.7±0.1 µm, having an aspect ratio around 86, were obtained. Some elementary fibrils (ca. 5 nm diameter) fibrillated from single BA nanofibrils were also observed (Figure 1b). Typically, single cellulose nanofibrils were reported to have a diameter of 5-50 nm and a length of few micrometers depending on processing conditions, sources, etc. [6].

![Figure 1](image1)

**Figure 1.** (a) SEM image of bagasse fibers (80x magnification) and (b) TEM image of nanofibrillated cellulose from banana pseudostem (8,000x magnification).

![Figure 2](image2)

**Figure 2.** SEM images of surfaces of bagasse sheets integrated with nanofibrillated cellulose (BA) at different contents. (a) 0 wt%; (b) 0.5 wt%; (c) 1.0 wt%; (d) 5.0 wt%.

SEM was used to examine the surface morphology of the pure bagasse sheet and the BA reinforced paper sheets. Micrograph obtained from the pure sheet (Figure 2a) revealed the large gaps/voids between the fiber network. Few fines (thin fibrils) were also observed in this bagasse sheet as
indicated with the black arrow. With increasing BA content, the presence of fines was raised confirming the retention of BA within the bagasse paper sheets (Figure 2b-d). These BA nanofibers were found to retain in the gaps between adjacent fibers, joining these fibers and also covered some parts of the fiber surfaces. Bridging of these fibers by BA creates a better bonding within the paper structures leading to a strengthening effect of the sheets [7,8]. This results are in good agreement with the increased mechanical properties of the reinforced paper sheets which are about to discuss in the following section.

![Mechanical properties of pure bagasse sheet (0%BA) and the sheets integrated with 0.5-5.0 wt% of nanofibrillated cellulose from banana pseudostem (0.5%BA-5%BA).](image)

**Figure 3.** Mechanical properties of pure bagasse sheet (0%BA) and the sheets integrated with 0.5-5.0 wt% of nanofibrillated cellulose from banana pseudostem (0.5%BA-5%BA).

From the tensile testing results (Figure 3), the reinforcing effect of BA was confirmed by the progressively improvement in the tensile index as well as the breaking length when the BA content in the paper sheets increased. Only 1.0 wt% BA addition could significantly increase both the tensile index (to 43 Nm/g) and breaking length (to 4377 m) of the paper sheet by 50%. The similar result has been recently reported by Espinosa et al. [3]. In their previous work, the incorporation of 1.5 wt% lignocellulosic micro/nanofibers (LCMN) from banana leaf residue was shown to enhance the breaking length of recycled papers from 2749 m to 3751 m (~36% increment) which became greater than the value requirement for using as fluting papers (> 3443 m).
Elongation is defined as the ability of the fiber network to stretch under load until breaking and is a good measure of the toughness of paper [8]. With increasing the BA content, the elongation of the current paper sheets increased considerably. This enhancement was believed to mainly come from a higher areas of inter-fiber bonding and bridging within the sheet structures by BA nanofibers as revealed by the previous SEM results (Figure 2). From the fractured surfaces comparing between the pure bagasse sheet (Figure 4a) and the 5.0 wt% BA integrated sheet (Figure 4b), differences in breaking mode of these two specimens were clearly seen. The failure with low degree of fiber stretching was observed in the pure bagasse sheet while, in the failed sheet with BA addition, yielding of the fibers and fines as well as slippage among them was noticed. Additionally, an increasing trend of the Young’s modulus (rigidity) of the sheets was also presented with higher BA contents (Figure 3).

4. Conclusions
Nanofibrillated cellulose from banana pseudostem (BA) was successfully prepared and used to reinforce in recycled bagasse paper sheets at 0.5, 1.0 and 5.0 wt%. SEM images revealed that BA nanofibers were physically retained on the sheet surfaces, bridging and connecting bagasse fibers together. This indicated an increase in fiber bonding areas within the sheet structures. All tensile properties of the BA reinforced sheets were noticeably enhanced, particularly their elongation or toughness. The BA integration led to a change in failure mode of the reinforced sheets allowing fibers to stretch and slide further before breaking. The current BA cellulose nanofiber demonstrated good potential as a reinforcing element for use in pulp and paper industry.

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