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Preparation of Pulp, Microfibrillated Cellulose, and Paper Hand Sheets from Bakong (*Hanguana malayana* (Jack) Merr.)

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ABSTRACT

Bakong (*Hanguana malayana* (Jack) Merr.) is a perennial herbaceous plant that grows abundantly in certain parts of the Philippines. This study investigates Bakong fibers' potential as a pulp source for papermaking. Furthermore, microfibrillated cellulose (MFC) produced from the fibers was used as an additive in the Bakong fiber-based hand sheets at 2%, 6%, and 10% w/w, and their effects on their strength properties were observed. The soda pulping method produced Bakong pulp with a total yield of 47.36%. Proximate chemical analysis showed that the method reduced the lignin content from 13.19% to 8.76%, and the resulting pulp was successfully formed into paper hand sheets. The strength properties of the hand sheets were tested, and the resulting burst index, tear index, tensile index, and folding endurance values were 5.98 kPa/m2·g, 6.41 mN·m2/g, 105.97 N·m/g, and 626 double folds, respectively. Aside from the tear index, the Bakong hand sheets' strength properties were much higher than that of locally produced commercial printing paper. The addition of MFC on the hand sheets negatively affected the burst and tensile index values of the Bakong hand sheets but significantly improved the tear index up to 6% w/w. These results show that the pulp produced from the Bakong fibers can potentially be used as an alternative source of pulp for papermaking, but further optimization of the pulping process is recommended to increase the yield, lower lignin content, and improve compatibility with the MFC.

Keywords: Microfibrillated cellulose, non-wood fibers, papermaking, pulping

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INTRODUCTION

The pulp and paper industry has been growing rapidly globally, so there is a great demand for raw pulp and paper materials. Since wood resources are scarce, there is increased interest in utilizing non-woody raw materials for pulp and papermaking (El-Sayed et al., 2020). *Hanguana malayana* (Jack) Merr., commonly known as Bakong, is a perennial herbaceous plant native to Sri Lanka, Southeast Asia, and Palau. In the Philippines, Bakong can be found in Laguna de Cagayan in Santa Teresita, Cagayan. It is an invasive species in the lake area, and excessive amounts may even be detrimental to the ecosystem (Choe et al., 2023).

As a fibrous plant, previous studies investigated utilizing its stems and leaves to produce fibers ranging from 1.2 to 1.4 m long. It has since been popular in the area and is projected to become a great source of livelihood for the people. According to the Design Center of the Philippines (2022), these fibers can be used as components for handicrafts and furniture designs. Other studies investigated its antibacterial use as leaf extractives are effective against certain bacteria (Borela et al., 2021).

However, not much research has been done on this plant and its utilization for pulp and papermaking. Thus, the potential of Bakong as a non-wood cellulose source is investigated in this study. Proximate chemical analysis was first performed to determine the holocellulose, lignin, and ash content of the Bakong fibers. Due to its similarities with other non-wood fiber sources, soda pulping was the chosen method to produce Bakong pulp (Wunna et al., 2017). The pulp produced from Bakong was processed to form hand sheets, and their strength properties were evaluated.

Moreover, this study also explored the use of Bakong pulp to produce nanocellulose. Since nanocelluloses were observed to increase the mechanical and barrier properties of paper, the Bakong pulp was further processed into microfibrillated cellulose (MFC) for use as an additive in the papermaking process (Balea et al., 2020). Using a friction grinder, MFC was produced by disrupting the intermolecular hydrogen bonding between the cellulose molecules in the Bakong pulp (Kargarzadeh et al., 2017). The MFC produced was then added to the Bakong hand sheets to explore its effect on the strength properties. A study by Viana et al. (2018) showed that adding up to 10% nanofibrillated cellulose positively affected the mechanical properties of paper hand sheets. Another study by Hollertz et al. (2017) showed that adding 2% chemically modified cellulose micro- and nanofibrils improved tensile strength when used as a paper additive. Based on these, this study investigated adding 2%, 6% and 10% MFC by weight.

MATERIALS AND METHODS

Proximate Chemical Analysis

Bakong stem fibers were received from the Design Center of the Philippines in connection with the Laguna de Cagayan Handicrafts Association. The fibers were bleached and decorticated in bundles approximately 1 m long. The fibers were washed with distilled water to remove dirt and other contaminants and then cut by hand to lengths of 50 mm. The composition of Bakong fibers was analyzed using the following methods: TAPPI T204 cm-07: Solvent extractives of Wood and Pulp, TAPPI T207 cm-08: water solubility of wood and pulp, TAPPI T211 om-02: Ash in wood, pulp, paper and paperboard, and TAPPI T222 om-02: Acid-insoluble lignin in wood and pulp. As a general baseline, the holocellulose content of the fibers was determined by difference.

Pulping and Handsheet Formation

In the study of Jimenez et al. (2005), the optimum pulping conditions for abaca, a non-wood plant with similar lignin content, were 170°C for 30 min, with 7.5% active alkali. Since the lignin content of the Bakong fibers is slightly higher, the pulping conditions were slightly modified. 600 g of air-dried 50 mm long Bakong fibers and 120 g NaOH in 4200 mL distilled water were cooked in a laboratory digester at 170°C for 2.5 h. The pulp was then washed with water until a neutral pH was achieved. The produced pulp was then passed through a disc refiner at 0.1 clearance, followed by beating using a Valley beater to achieve pulp freeness values of 250 Canadian Standard Freeness (CSF).

A standard 159 mm diameter sheet machine with a stirrer produced Bakong hand sheets based on the TAPPI Standard T205. A 30 g of Bakong pulp was diluted and disintegrated at 3000 rpm for 10 min. The slurry was then fed to a TAPPI hand sheet former cylinder to produce 80 g/m^2 hand sheets. The hand sheets were pressed for 10 min at 345 kPa and dried at 27°C for 24 h. The hand sheets were then conditioned in an atmosphere with a temperature of 23±1°C and 50±2% relative humidity before testing.

Preparation of Bakong Microfibrillated Cellulose (MFC)

Bakong pulp was first milled to 2 mm lengths to increase surface area for the bleaching process. Hypochlorite bleaching was performed using a modified method based on a previous study (Yuanita et al., 2015). The pulp was then soaked in 5.25% sodium hypochlorite solution at 10% w/v consistency for 30 min at room temperature. The bleached pulp was washed with distilled water until a neutral pH was achieved. The bleached pulp was soaked with distilled water at 1% w/v and subjected to a mechanical friction grinding process using the Masuko Supermasscolloider, model MKCA6-2. The grinder was operated at a speed of 1500 rpm, and the suspension was subjected to grinding for 15 passes. The grinder clearance was reduced every 1 to 2 passes until a clearance of -10 was reached at the 15th pass. The produced MFC was incorporated into the hand sheets at a 2%, 6% and 10% w/w ratio before formation.

A gravimetric test was performed to test for the retention of MFC in the hand sheets. Hand sheets were made from pulp with known oven-dry weights. Hand sheets with additional MFC of known oven-dry weights were also prepared. The hand sheets were oven-dried at 105°C and weighed every hour until the weight variation was less than 0.01 g. Afterward, the weights of the hand sheets with and without MFC were compared.

Characterization of Produced Hand Sheets

Surface Morphology

A scanning electron microscope was used to capture high-magnification micrographs of the surface of the Bakong hand sheets. Due to their organic and non-conductive nature, the samples were ion-coated prior to viewing under the microscope.

Handsheet Testing

The grammage of the produced hand sheets was determined using ISO standard 534. The hand sheets were cut according to TAPPI T220 to test the mechanical properties. The hand sheets' burst index, tensile index, tear index, and folding endurance were tested according to ISO standards 2758, 1924-2, 1974, and 5626, respectively. Five replicates were done for each type of hand sheet produced. For comparison, commercial-grade paper was tested using the same methods. The data obtained were subjected to a one-way analysis of variance (ANOVA) at a 95% confidence level ($p \leq 0.05$) to check if the differences among the compared groups were statistically significant. It was followed by the Tukey-Kramer post hoc test to analyze the differences when significance was observed.

RESULTS AND DISCUSSIONS

Chemical Composition of Bakong Fibers

The Bakong fibers received were air-dried and brown. Milling was performed using a Wiley mill to reduce the size of the fibers to about 2 mm long. (Figure 1). A summary of the chemical composition of the Bakong fibers based on the oven-dry weight is shown in Table 1. The moisture content of the Bakong samples after air-drying for at least 24 h was found to be 13.12%. It was the basis for determining the chemicals needed for the succeeding proximate analysis and pulping.

Figure 1. Bakong fibers: (a) as received, (b) after milling, (c) after hand sheet formation

Table 1
Chemical composition of Bakong fibers

Chemical Components	Compositions (%)
Moisture Content	13.12 ± 0.45
Ash Content	7.09 ± 0.10
Cyclohexane-ethanol Extractives	2.25 ± 0.09
Hot Water Extractives	8.78 ± 0.20
Lignin Content	13.19 ± 1.16
Holocellulose Content (by difference)	68.69 ± 2.00

The fibers' ash content was 7.09%, which is typical of wheat straw species (Plazonic et al., 2016). It is considered relatively high and may be disadvantageous in pulp and papermaking as they could interfere with pulping and recovery of pulping chemicals. The cyclohexane-ethanol extractives and hot water extractives of the Bakong fibers were 2.25% and 8.78%, respectively. These values add up to 11.03%, much higher than wood's, typically containing 3 to 8% of total extractives and normally not exceeding 5% (Lehr, 2021). These negatively affect the pulping and bleaching processes, as they can react with the pulping liquor and block the diffusion of bleaching chemicals and lignin (Dai, 2001). It is suggested that additional pre-treatment methods be employed to lower the ash and extractive content of the fibers before they are used in pulping and papermaking.

The lignin content of the fibers was found to be 13.19%, which is generally lower than those of wood species. Additionally, the holocellulose content of the fibers was found to be 68.69%. It shows that Bakong is a promising source of pulp, having similar holocellulose content values compared to other fiber sources (Moral et al., 2017). Soda pulping was chosen since the raw material contains significantly lower lignin than wood sources. Moreover, Kraft pulping was not employed to minimize the use of sulfur-containing compounds, which are considered environmental pollutants.

Production of Bakong Pulp and Microfibrillated Cellulose

The soda pulping yield of the Bakong fibers is listed in Table 2. On average, the pulping method yielded 48.85% yield, with the resulting pulp having a lignin content of 8.76%. It is within the range of typical Kraft pulping yields, which is between 43 to 70% (Biermann, 1996). However, this value is much lower than that obtained by Jimenez et al. (2005) for abaca, which is 77.33% and has a lignin content of around 4.8%. Although the method produced pulp from the Bakong fibers with a reasonable yield and removed 70% of the lignin content of the original fibers, improvements can still be made to the process.

The pulp underwent refining and beating to further increase the fiber surface area and make it more suitable for papermaking. A beating time of 5 min, with a 5 kg load, was determined for the pulp to reach freeness values of around 250 CSF, which is typical for paper stock. Circular hand sheets were successfully produced after the beating process, as

Table 2
Soda pulping and refining yield of Bakong fibers

Process	Yield(%)
Soda Pulping, Beating, and Refining	47.36 ± 2.89
Pulp Bleaching	66.47 ± 3.10
Friction Grinding (Masuko Supermasscolloider)	87.53 ± 0.056
Overall MFC yield	27.5 ± 2.98

shown in Figure 1, with minimal losses. The overall yield after beating was determined to be 47.36%, showing that Bakong can be a good non-wood alternative source of pulp for papermaking.

To produce MFC, the Bakong pulp was bleached to reduce lignin content to as low as 1.42%. After friction grinding using a Masuko Supermasscolloider, the resulting product is a white suspension with a gel-like consistency. The bleaching and friction grinding yields are summarized in Table 2. An overall yield of 27.55% was obtained, and this value may be attributed to the low yields after pulp bleaching. It suggests that the bleaching process degraded the celluloses and hemicelluloses in the fibers, and further optimization of the process is recommended.

The MFC was incorporated into the hand sheets by adding the suspension into the pulp before formation. MFC retention was determined to be 94.4%. Thus, the method used successfully incorporated the MFC into the hand sheets.

Microscopy of the MFC and Hand Sheets

The formed hand sheets were observed under a scanning electron microscope. The pulped Bakong fibers were randomly oriented within the hand sheet. The fibers had diameters of 20 μm and lengths greater than 2 mm, as shown in Figure 2. Destruction of natural fiber bundles during the bleaching and friction grinding process resulted in fibrillation. In a previous study, alkali and oxidative bleaching of fibers reduced fiber diameters from 75–90 μm to 5–6 μm (Yuanita et al., 2015). In this study, while milder bleaching conditions were

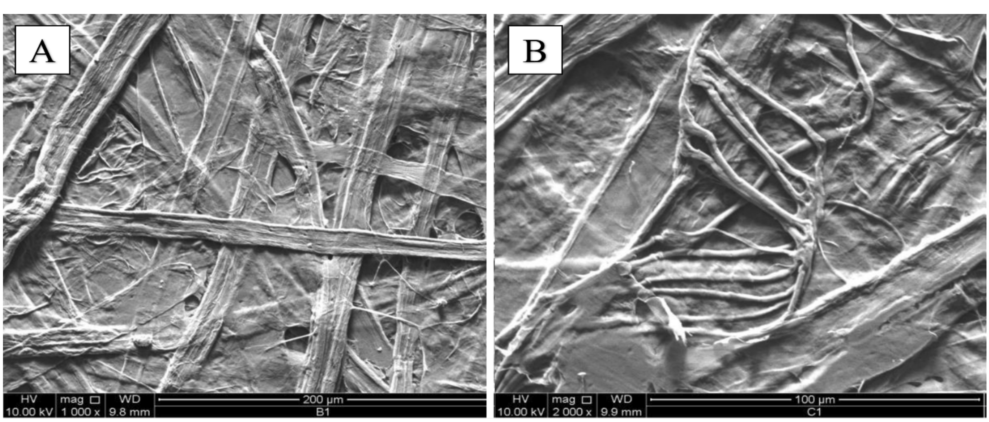

Figure 2. SEM images of the hand sheet (a) without MFC and (b) with MFC

used, friction grinding was employed to further fibrillate the Bakong fibers. Under SEM, the fiber diameters were reduced to about 3 μm. The hand sheets with additional MFC showed the presence of these separate smaller fibers on the surface, which were presumed to be MFC adhered to the pulp fibers.

Bakong Pulp Handsheet Properties

Hand sheets were produced from the Bakong pulp with grammage values of 80 g·m^{-2}. A summary of the strength properties of the produced hand sheets is shown in Table 3. Commercial printing paper produced locally (labeled "C") was also tested using the same methods and shown for comparison. The values listed for the commercial printing paper were for the machine direction, which are normally higher than those for cross direction. The effect on the strength properties of adding MFC to the Bakong pulp during hand sheet formation was also tested. Figures 3–6 show the burst index, tear index, tensile index and folding endurance, respectively.

Table 3
Mechanical properties of hand sheets produced from Bakong pulp compared to commercial printing paper

Mechanical Properties	Bakong Hand Sheets Prepared in This Study	Commercial Printing Paper
Burst Index (kPa·m^{-2}·g^{-1})	5.98 ± 0.23	3.01 ± 0.13
Tear Index (mN·m^{2}·g^{-1})	6.41 ± 0.55	7.53 ± 0.18
Tensile Index (N·m·g^{-1})	105.97 ± 3.97	52.56 ± 2.19
Folding Endurance (double folds)	626 ± 189	41 ± 8.29

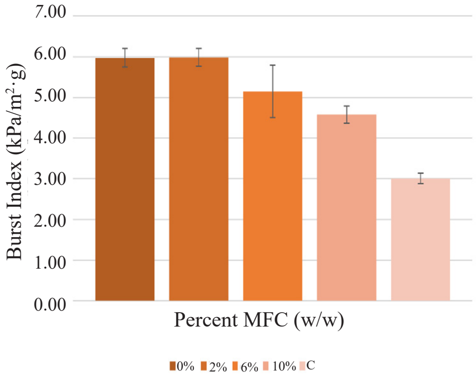

Figure 3. Burst index of the produced hand sheets

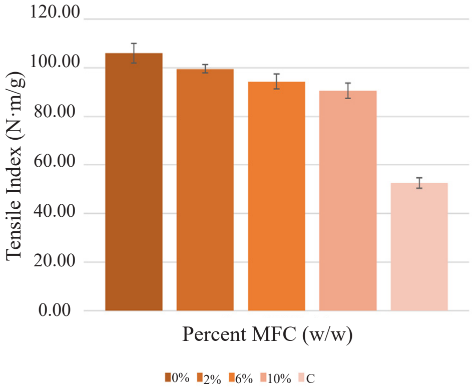

Figure 4. Tensile index of the produced hand sheets

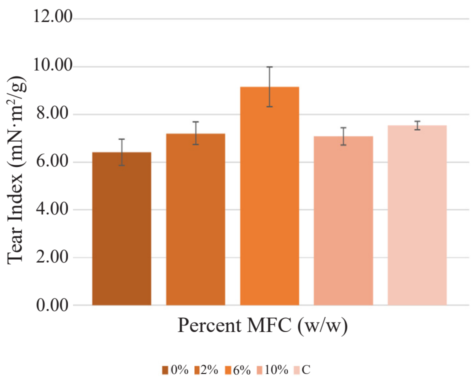

Figure 5. Tear index of the produced hand sheets

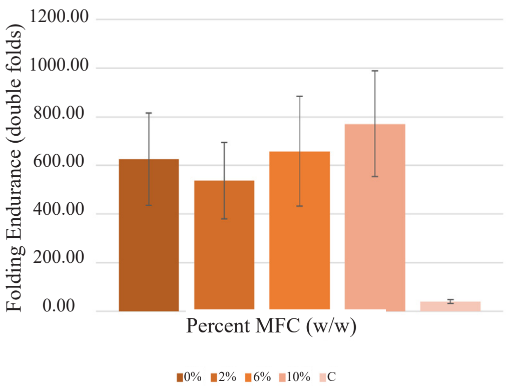

Figure 6. Folding endurance of the produced hand sheets

The burst and tensile indices of the hand sheets were tested as general indicators of their strength characteristics (PITA Raw Materials Working Group, 2005). It was observed that the hand sheets produced from the Bakong pulp had significantly greater burst and tensile strength than that of commercially produced printing paper. It could be attributed to the long fibers' strong inter-fiber bonding. It shows that the pulp suits withstanding strains associated with the papermaking process and end-use.

However, with the addition of MFC, the burst index and tensile index values decreased noticeably. A similar case was found in the study of Viana et al. (2018), wherein no significant improvement in the mechanical property of newsprint was observed with the addition of nanofibrillated cellulose. Since the Bakong pulp fibers still contained a significant amount of lignin, adding MFC may have been less compatible with the fibrous elements and did not promote inter-fiber bonding.

A different trend was observed when testing the tear index of the formed hand sheets. Although tear resistance also depends on the degree of bonding between the fibers, it mostly depends on the fiber elements' resistance, like the wall's length and thickness (Viana et al., 2018). The addition of up to 6% MFC reinforced the thickness of the fiber walls by bonding with the surface. Further increasing this to 10% resulted in weaker inter-fiber bonding, ultimately negatively affecting the tear index. Since the tear index of Bakong hand sheets was much lower than that of commercial printing paper, they may not be suitable for wrapping or packaging applications. However, the tear resistance was improved significantly with the addition of 6% MFC. Although the other mechanical properties decreased with the addition of MFC, at a level of 6% by weight, all the mechanical properties of the MFC-reinforced Bakong hand sheets were still higher than those of commercial printing paper.

Lastly, the folding endurance of the fibers was tested. The test gives extremely variable results. Using one-way ANOVA, no significant property differences were observed between the untreated hand sheets and those reinforced with MFC (p-value > 0.05). However, the folding endurance of the Bakong hand sheets was much higher than that of printing paper. Thus, Bakong pulp can be an additive for specialty papers requiring high folding endurance, such as currency bank paper. Along with Abaca, Salago and other fibers, it can contribute to the country's goal of producing banknotes using 100% locally sourced fibers. (Fernandez, 2018).

CONCLUSION

Pulp, microfibrillated cellulose, and paper hand sheets were produced from Bakong (*Hanguana malayana* (Jack) Merr.) fibers. Soda pulping at 170°C and 7.5% active alkali effectively isolated the individual plant fibers. However, additional pre-treatment methods are recommended for commercial pulping purposes due to the plant material's high ash content. The pulping yields were high, but the pulp still had a considerably high lignin content, so optimization of the process is needed. Bleaching conditions using 5.25% sodium hypochlorite were effective in further lignin removal, which was necessary for producing microfibrillated cellulose (MFC) and using friction grinding. MFC yields were 87.53%. Scanning electron microscopy supported that microfibrillation occurred and that the MFC had fiber widths of 3 µm. The MFC produced was successfully incorporated into the hand sheets made from Bakong pulp. The MFC was added at 2, 6, and 10% w/w, and it was observed that the burst and tensile indices of the pulp slightly decreased with increasing MFC loading. However, the tear index was greatly improved, and compared to commercial printing paper, the MFC-reinforced Bakong pulp had desirable properties at 6% w/w loading. Bakong pulp is a suitable candidate for use as an alternative to wood pulp in papermaking. Moreover, MFC from Bakong pulp may be used as an additive to commercial pulp to improve its tear resistance, but this requires further study. Overall, the Bakong fibers showed promise as a source of pulp for both microcellulose production and papermaking. The MFC yields were relatively high, and the hand sheets had good mechanical properties.

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