**A Study on the Use of Food-Grade and Non-Food Grade Reagents in the Extraction of Cellulosic Fibers from Bamboo Species in Baguio City, Philippines**

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**Abstract.** Cellulosic fibers were extracted from bolo (***Gigantochloa bolo***) culm and running bamboo (***Phyllostachys aurea***) branches grown in Baguio City, Philippines using separate pre-treatment and processing reagents. Food grade reagents such as acetic acid and hydrogen peroxide were used to further process the fibers after NaHCO3 pre-treatment and non-food grade reagents such as hydrochloric acid were used to further refine the NaOH pre-treated fiber samples. Fourier Transform Infrared (FTIR) analysis revealed that the use of food grade reagents in the fiber extraction process was effective in removing surface impurities as shown in the general decrease in percentage absorbance (%A) of functional groups associated to hemicellulose and lignin. This is also observed in the increasing crystallinity index (CI) calculated from x-ray diffractogram for six days NaOH pre-treatment, six days NaHCO3 pre-treatment, and 10 days NaHCO3 pre-treatment of bolo culms. However, in terms of fiber diameter NaOH pre-treatment was best in decreasing fiber diameter in running bamboo stem as shown in the optical microscopy results. The scanning electron microscopy (SEM) displayed a smoother and more organized structure for NaOH pre-treated bamboo fibers. In general, volumetric analysis using potassium dichromate (K2Cr2O4) and ferrous ammonium sulfate (FeH8N2O8S2) in a reduction-oxidation reaction confirmed the presence of alpha(α) cellulose in the bamboo samples as evidenced by a purple end-point. Further investigation of processing parameters using food grade or eco-friendly reagents can be done as this initial investigation shows the potential of food-grade reagents in the extraction of cellulosic fibers from bamboo.

**Keywords:** Cellulosic Bamboo Fibers, Alkaline Pre-Treatment, Food Grade Reagents

1. Introduction

The usual process of fiber extraction from plants includes mechanical crushing, steam explosion techniques, use of biological enzymes and water retting techniques. Chemical pre-treatment methods such as use of NaOH, HCl, H2SO4 are also considered in combination with mechanical crushing or steam explosion. While chemical pre-treatment methods are proven to be effective, the reagents are toxic to human health and environment with large-scale use. The use of food grade reagents such as sodium bicarbonate (NaHCO3) and acetic acid (CH3COOH) in vinegar are cheaper and safer to work on. These are also widely available and easy to purchase.

The use of ecofriendly reagents such as ethanol, acetic acid, and hydrogen peroxide was done by Rehman *et al.* in 2018 and they were able to successfully isolate cellulose from eucalyptus plant with 73% crystallinity [1]. 12 wt% of NaHCO3 treatment for 120 hours soaking time on coir fibers were also performed. The resulting fibers increased in tensile strength, observance of lesser micropores thus decreased local stress, and increased crystallinity index [2]. NaHCO3 treatment was also reported to be effective and efficient in removing the surface impurities and hemicellulose content of jute/hemp/flax bidirectional mats. 9% of NaHCO3 solution with 96-hour soaking time was concluded to be effective [3]. Untreated Napier grass fiber strand was also compared to acetic acid treated samples. Acetic acid at 15 wt% and 2 hours soaking time was found to have increased crystallinity index and thermal stability of the fiber samples [4]. In another study, 20% NaHCO3 solution at 120 hours soaking time was effective on flax fiber extraction. The extracted fibers’ tensile strength was reported to increase tensile strength by 43% and increased young’s modulus at 81% [5]. Fiber treated with NaHCO3 when used to reinforce polymer composites resulted to better properties. The tensile and flexural properties of polymer composites reinforced with flax, jute, and sisal fibers were also observed to show significant increase as compared to composites reinforced with NaOH treated fibers [6]. The study made by dos Santos *et al.* has also shown that laminates made with polyester polymer and sisal fiber treated with Na2CO3 for 96 hours have remarkable increase in tensile and flexural modulus as well as reduced impact resistance [7].

 The extraction of fibers from bamboo were also experimented on due to bamboo fibers’ potential use in manufacturing industries. Bamboo fibers can replace petrochemical synthetic fibers due to its superior mechanical properties, sustainability and versatility. Zheng *et al.* used the acetic acid ball milling technology as pre-treatment to moso bamboo powder to substitute traditional fiber board and particle board as furniture materials. The acetic acid ball-milled moso bamboo powder was more compact resulting to a superior mechanical property in terms of tensile and specific strength [8]. Steam explosion, mechanical techniques, and chemical extraction using alkali or NaOH were used to extract bamboo fibers. It was found that the chemical treatment increased the interfacial shear strength (IFSS) of bamboo with polyester and polypropylene composites. It was suggested by the researchers that airing of waste water from the alkaline extraction process using CO2 gas can reduce the pH of waste water from 10.8 to 7.0 within 110 minutes aeration time [9]. In another study, it was also discovered that alkali or NaOH treatment could remove lignin from bamboo fibers thus improving the interfacial adhesion of bamboo fibre-reinforced composites [10].

This study on the use of eco-friendly reagents to extract fibers from bolo (***Gigantochloa bolo***) culm and running bamboo (***Phyllostachys aurea***) branches would be an ecologically friendly means of fiber extraction. The inorganic reagents sodium hydroxide (NaOH), sulfuric acid (H2SO4), and hydrochloric acid (HCl) used in the processing of plant fiber sources for extraction purposes are both corrosive and toxic to humans who are often exposed to these. The disposal of waste water from the extraction process poses problem to the environment. The use of food grade reagents such as sodium bicarbonate (NaHCO3) /baking soda and acetic acid (CH3COOH) in vinegar would be safer to handle and dispose aside from its availability. If proven to be effective, then bamboo fibers extracted using food grade reagents can be incorporated in food bio- packaging applications, edible bioplastics, textile and composite fabrication in industries - aeronautics, automotive, furniture. However, the mechanical properties of extracted fibers, applications in composite and textile fabrication still need to be explored and experimented on as a result of this groundwork study.

1. Materials and Methodology

Two species of bamboo, the bolo (***Gigantochloa bolo***) culm and running

bamboon(***Phyllostachys aurea***) branches were used to extract fibers. These are locally grown in Baguio City, Philippines.

**2.1 Bamboo Fiber Extraction Process and Characterization Techniques**

 Five to seven grams of chopped and sun-dried bamboo samples from bolo culms and running bamboo branches were pre- treated with 1 M NaOH for 6 days, mechanically crushed in a mortar and pestle then acid hydrolyzed with 1 M HCl at 70 °C - 80 °C for 2 hours. The acid hydrolyzed samples were again soaked in 1 M NaOH for 2 hours at 70 °C – 80 °C followed by hydrogen peroxide treatment, 30% v/v solution, for 12 hours. The chemically treated fibers were subjected to a high-pressure defibrillation for 1 minute using an Oster osterizer to separate the lignin, hemicelluloses, and the crystalline portion of the fiber. The fiber extraction of five using food grade reagents employed the same method stated using food grade reagents such as sodium bicarbonate (NaHCO3), acetic acid (CH3COOH), and hydrogen peroxide (H2O2).

The characterization techniques employed were **(1)** Fourier Transform Infrared with Attenuated Total Reflectance (FTIR-ATR) analysis using Agilent Cary 630 FTIR was done to determine the percentage absorbance (%A) of functional groups at 890-900 cm-1 due to *ᵦ*- glycosidic linkage by both cellulose and hemicellulose [11];

**(2)** The Scanning Electron Microscope (SEM) used was JEOL JSMIT500HR/LA. It was employed in the study to further understand the surface morphology of the extracted fiber strands from bamboo fiber. The samples were coated with platinum and processed using a landing voltage of 15.0 kV at 1,000 X magnification under high vacuum; **(3)** X-ray diffraction (XRD) analysis was performed to know the Crystallinity Index (CI) of the extracted bamboo fibers.TheShimadzu Lab X XRD-6100 X-ray Diffractometerwas the brand of the machine, programmed at θ Range: 5 o to 60 o with the rate of 1.0 o per minute**.** Fiber samples were pressed in the sample holder. The CI was calculated from the XRD data using the Segal Method [12]; **(4)** Optika compound microscope, B-190 series at 50X magnification was used to determine fiber fineness. The external fiber diameter was measured using a microscope micrometer placed on the stage. Twenty random fiber strands were picked and placed on the stage for measurement. **(5)** Bamboo fibers was reported to contain 40% - 50% α cellulose [13]. In the study, the extracted bamboo fibers were qualitatively tested of its α cellulose content using a reduction-oxidation reaction of potassium dichromate (K2Cr2O4) and ferrous ammonium sulfate (FeH8N2O8S2). The procedure was adopted from Technical Association of the Pulp and Paper Industry, TAPPI,1999 [14].

1. Results and Discussion

The extraction of bamboo fibers from culm of bolo (***Gigantochloa bolo***) and branches of running bamboo (***Phyllostachys aurea***) were successfully performed using 6 days NaOH pre-treatment followed by additional procedures described in the methods portion of this paper which is identified as use of non-food grade reagents. Apparently, the 6 days and 10 days NaHCO3 pre-treatment followed by extraction procedures identified as use of food grade reagents were also observed to be a success except for some processing parameters that need further improvement.

 Thorough inspection of the FTIR spectra reveals an increase in the percentage absorbance (% A) of hydroxyl groups, lignin, and glycosidic linkage by both cellulose and hemicellulose for 6 days NaOH pre-treatment and 6 days NaHCO3 pre-treatment. The OH groups contributed by cellulose, hemicellulose, and lignin did not decrease with NaHCO3 pre-treatment which means that surface impurities were not removed by NaHCO3 pre-treatment for 6 days. Yet, at 10 days NaHCO3 pre-treatment time there was a decrease in the % A of OH groups and β-glycosidic linkage by cellulose and hemicellulose. This clearly explains the need for longer NaHCO3 pre-treatment time of the bamboo culms (***Gigantochloa bolo***) to effectively remove surface impurities such as lignin and hemicellulose thereby extracting more cellulose from the fiber bundle. It is also suggested to chop the bamboo culm (***Gigantochloa bolo***) into thinner pieces so that NaHCO3 will easily penetrate into the fiber bundles that will eventually cause removal of hemicelluloses and lignin.

 In plants, the fiber bundle functions for structural support making the plant stronger. When fibers are used as a material in composites and textiles, hemicelluloses and lignin are weaker and decrease interfacial adhesion to other bio-polymers therefore the more crystalline cellulose is of interest to material scientists. The use of a basic solution such as NaOH and NaHCO3 in fiber pre-treatment breaks the hydroxyl (OH) bonds that holds cellulose, hemicelluloses, and lignin together. Depolymerization occurs, producing a more crystalline fiber [15].

A general decrease in the % A of functional groups related to cellulose, hemicelluloses, and lignin for running bamboo (***Phyllostachys aurea***) branches from 6 days NaOH pre-treatment to 6 days NaHCO3 and 10 days NaHCO3 pre-treatment was observed. The decrease in %A means that the 6 days and 10 days pre-treatment time for running bamboo (***Phyllostachys aurea***) branches was effective in removing hemicelluloses and lignin from the fiber bundle, thus more cellulose is obtained. While NaOH pre-treatment is proven to be good in extracting cellulosic fibers, the best pre-treatment time with the use of NaHCO3 on running bamboo (***Phyllostachys aurea***) is 10 days exemplified by the remarkable decrease in %A of functional groups related to cellulose, hemicellulose, and lignin.

SEM analysis reveals a more fibrous structure for NaOH pre- treated bolo (***Gigantochloa bolo***) culm and running bamboo (***Phyllostachys aurea***) fibers. The 6 days pre-treatment using 1 M NaOH for both bamboo culm (***Gigantochloa bolo***) and running bamboo (***Phyllostachys aurea***) branches was effective for producing a more fibrous and less damaged bamboo fiber. Bolo culm fiber pre-treated with 8 wt % NaHCO3 for 6 days and 10 days have torn and rougher structure. This might mean that the pre-treatment time and concentration might not be enough to produce a more fibrous structure. However, for running bamboo (***Phyllostachys aurea***) branches the 6 days pre-treatment time with NaHCO3 starts to produce more longitudinal structure and at 10 days pre-treatment fibers are being separated from the fiber bundle. This means that NaHCO3 pre-treatment is effective in separating the bamboo fiber bundles into fiber strands. Perhaps a longer pre-treatment time is still needed to produce a finer fiber structure.

X-ray diffraction analysis gives an idea of the crystallinity index (CI) of the extracted fibers. It was seen that the most crystalline fiber was extracted from running bamboo (***Phyllostachys aurea***) branches pre-treated with NaOH at 6 days soaking time. For bolo (***Gigantochloa bolo***) culm, the most cellulosic fiber came from the 10 days NaHCO3 pre-treated sample.

Fiber fineness is measured using the external diameter of the extracted fiber using an optical microscope. The fiber diameter on the average, 78.75 ± 70 microns for extracted bolo (***Gigantochloa bolo***) culm fibers were finer for NaOH pre-treated sample. This is similar to the extracted running bamboo (***Phyllostachys aurea***) fiber using NaOH pre-treatment with an average fiber diameter of 17.88 ± 14. However, it can be seen that longer pre-treatment time for NaHCO3 soaked running bamboo (***Phyllostachys aurea***) is needed to improve fiber fineness since the diameter of the 6 days pre-treated sample and 10 days pre-treated sample decreased from 70.38 ± 84 to microns to 52.00 ± 87 microns. This is similar to the work done on alkali pre-treated kenaf fibers where increasing soaking time causes reduction in fiber diameter due to the degradation of impurities such as lignin with alkalinization [16]. Comparing the external fiber diameter of extracted bamboo fibers in this study to the study done by other researchers with a value of 107.9 um using similar pre-treatment [17], it can be said that most of the extracted fibers in this study are finer at 78.75 um,17.88 um, 70.38 um, and 52.00 um.

Alpha (α) cellulose is the undegraded form of cellulose using volumetric method of analysis. The color change at end-point for a positive result is from brown to purple. TAPPI procedure for alpha cellulose extraction was done and the bamboo fiber samples from bolo (***Gigantochloa bolo***) culm and running bamboo (***Phyllostachys aurea***) branches gave a purple end-point after the titration process. This signifies the presence of α – cellulose from the extracted bamboo fibers.

1. Conclusion

While sodium hydroxide (NaOH) pre-treatment and the use of non-food grade reagents are effective in the extraction of a more crystalline and finer bamboo fibers, the use of food grade reagents such as sodium bicarbonate (NaHCO3), acetic acid (CH3COOH) in vinegar, and food grade hydrogen peroxide (H2O2) can also produce a comparable result when processing parameters are improved. It can be suggested that longer-pre-treatment time and processing time using food-grade reagents be explored to separate a more crystalline and cellulosic fiber from the bamboo fiber bundle. Bamboo samples should be chopped into smaller size before NaHCO3 pre-treatment to allow lignin, a component of the fiber bundle depolymerize and solvate in the alkaline environment. Depolymerization occurs when bonds at β-O-4 (aryl-glyceryl-β-aryl ether bond) and 4-O-5 bond (diaryl ether bond) break. In the same way, smaller oligomers and xylose are also produced in a basic environment by hydrolysis of hemicelluloses. It is therefore concluded that the use of food grade reagents is possible in the extraction of cellulosic bamboo fibers from bolo culm and running bamboo to be used in food bio- packaging applications, edible bioplastics, textile and composite fabrication in industries - aeronautics, automotive, furniture.

References

1. Rehman, N.: Alam, S.: Ul Amin, N.: Mian,I.; Ullah,H. Ecofriendly Isolation of Cellulose

from Eucalyptus lenceolata: ANovel Approach. International Journal of Polymer Science, 2018 (1), 1-7 (2018).

2. Bakri, B.: Putra, A. E. E.: Mochtar, A. A.: Renreng, I.: Arsyad, H. Sodium bicarbonate treatment

 on mechanical and morphological properties of coir fibres. International Journal of Automotive

and Mechanical Engineering, 15(3), 5562-5572 (2018).

3. Das, P. P.: Manral, A.: Ahmad, F.: Sharma, B.: Chaudhary, V.: Gupta, S.: Gupta, P.

Environmentally sustainable chemical treatment of plant fibers for improved performance of polymeric composites. Polymer Composites, 43(10), 7155-7169 (2022).

4. Parasuram, K. V.: Reddy, K. O.: Shukla, M.: Marwala, T. Morphological, structural and thermal

characterization of acetic acid modified and unmodified napier grass fiber strands. In: 2013 7th International Conference on Intelligent Systems and Control (ISCO), pp. 506-510. IEEE Xplore, Canada (2013).

5.Belaadi, A.: Amroune, S.: Bourchak, M. Effect of eco-friendly chemical sodium bicarbonate treatment

on the mechanical properties of flax fibres: Weibull statistics. Int J Adv Manuf Technol 106, 1753–1774 (2020).

6.Benkhelladi, A.: Laouici, H.: Bouchoucha, A. Tensile and flexural properties of polymer

composites reinforced by flax, jute and sisal fibres. The International Journal of Advanced Manufacturing Technology 108, 895-916 (2020).

7.Santos, J. C. D.: Oliveira, P. R.: Freire, R. T. S.: Vieira, L. M. G.: Rubio, J. C. C.: Panzera, T. H. The

effects of sodium carbonate and bicarbonate treatments on sisal fibre composites. Materials Research, 25, e20210464 (2022).

8. Zheng, G.: Ye, H.: Liang, Y.: Jin, X.: Xia, C.: Fan, W.: Xie, Y.: Shi,Y.: Li,J.: Ge, S. Pre-treatment of natural bamboo

for use as a high-performance bio-composites via acetic acid ball milling technology. Construction and Building Materials 367, 130350 (2023).

9. Phong, N. T.: Fujii, T.: Chuong, B.: Okubo, K. Study on how to effectively extract bamboo fibers from

raw bamboo and wastewater treatment. Journal of Materials Science Research, 1(1), 144 (2012).

10. Zakikhani, P.: Zahari, R.: Sultan, M. T. H.: Majid, D. L. Extraction and preparation of

 bamboo fibre-reinforced composites. Materials & Design 63, 820-828 (2014).

11. Mortazavi, S. M.: Moghadam, M. K. Introduction of a new vegetable fiber for textile

 application. Journal of Applied Polymer Science, 113(5), 3307-3312(2009).

12. Segal L.: Creely J.J.: Martin A.E.: Conrad C.M. An Empirical Method for Estimating the Degree of Crystallinity

 Native Cellulose Using the X-Ray Diffractometer. Textile Research Journal. 29(10),786-794 (1959).

13. Nayak, L.: Mishra, S.P. Prospect of bamboo as a renewable textile fiber, historical overview, labeling,

 controversies and regulation. Fash Text 3, 2 (2016).

14. T 203 cm-99 TENTATIVE STANDARD. Approved by the Chemical Properties Committee of the Process

and Product Quality Division TAPPI. <https://www.tappi.org/globalassets/t429-d-2-b-3-sarg.pdf> (1999), last accessed 2024/10/01.

15. Mwaikambo, L.Y.: Ansell, M.P. Chemical modification of hemp, sisal, jute, and kapok fibers

 by alkalization. J. Appl. Polym. Sci. 84, 2222-2234 (2002).

16. Hashim,M.Y.: Amin,A.M.: Marwah,O.M.F.: Othman,M.H.: Yunus,M.R.M.: Huat,N.C. The effect of alkali

treatment under various conditions on physical properties of kenaf fiber. In: Journal of Physics: Conference Series 914 012030. IOP Science, England (2017).

17. Hu,M.: Wang,C.: Lu,C.: Anuar,N.I.S.: Sheraz, H.S.Y.: Jing,M.: Chen,Z.: Zakaria,S.: Zuo,H. Investigation on the

 Classified Extraction of the Bamboo Fiber and Its Properties. Journal of Natural Fibers, 17(12), 1798-1808, (2020).

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