Biointerface Research in Applied Chemistry
Open-Access Journal (ISSN: 2069-5837)

Article
Volume 13, Issue 5, 2023, 444
https://doi.org/10.33263/BRIAC135.444

Characterization of some Brazilian Lignocellulosic Materials for Nanocellulose Production

Heloise Sasso Teixeira [1,2], Marina B. Fogaça [3], Fabio Furtado [3], Marcia R. Leal [3], Aline Caldonazo [1], Talita Szlapak Franco [4], Thais H. Sydenstricker Flores-Sahagun [1,3,*], Alan S. Andrade [4], Milton O. Vazquez-Lepe [5], Graciela I. Bolzon Muniz [4]

1 Post-graduation Program in Engineering and Materials Science/PIPE, Federal University of Parana, Curitiba, 81531-980, Brazil
2 Post-graduation Program in Materials Science, University of Guadalajara, Guadalajara, Jalisco, 44430, Mexico
3 Post-graduation Program in Mechanical Engineering, Federal University of Paraná (UFPR), P.O Box 19049, 81531-990, Curitiba, Paraná, Brazil
4 Post-graduation Program in Forest Engineering, Federal University of Paraná (UFPR), Curitiba, Av Prefeito Lothário Meissner, 632, Curitiba, Paraná, 80210-170, Brazil
5 Department of Project Engineering, University of Guadalajara, Guadalajara, Jalisco, 45100, Mexico
* Correspondence: tsydenstricker@gmail.com (T.H.S.F.-S.);

Scopus Author ID 36677293700
Received: 4.09.2022; Accepted: 7.10.2022; Published: 21.12.2022

Abstract: Considering the importance of the use of alternative vegetable fibers in the production of nanocellulose, in this work, four Brazilian fibers, Pampas-grass, silk-floss, coconut, and *Phormium tenax* were chemically, morphologically, and thermally characterized. Composition, solubility extractives, and crystallinity indexes of fibers indicate how aggressively or mildly the fiber treatment may be carried out to produce nanofibers. The use of coconut fibers stored for a long time (17 years) resulted in fibers with much lower lignin content, which may be interesting considering other lignocellulosic fibers. According to the specific characteristics of each fiber, chemical and/or mechanical treatments were applied. The one that resulted in the lowest lignin content was chosen to produce nanocellulose characterized by scanning (SEM) and transmission (TEM) electron microscopy. SEM microscopy of powder silk-floss nanocellulose was also shown without any chemical treatment.

Keywords: Brazilian fibers; nanocellulose; nanofiber; cellulose; lignocellulose.

1. Introduction

The need to replace polymers derived from non-renewable sources with those from renewable and biodegradable sources (eco-friendly bioplastics) has increased the use of lignocellulosic biomass, which comprises the most abundant source of polymers in nature [1-5]. Polylactic acid, polyhydroxyalkanoates, bio-polyethylene (HDPE and LLDPE), polyurethanes, and starch-based nanocellulosic bioplastics may be derived from lignin or cellulose [6]. Among other factors that increase its competitiveness and interest, bioplastics' relevance lies in their relatively low cost and ability to replace oil derivatives in different sectors such as packaging, formulation of composites, biomedical devices, healthcare, cosmetics, electronic, automotive products, textiles, optical devices, among others [7-11].

The nanometric portion of cellulose has even greater potential, and its extraction from different sources is extensively reported [12-14]. In addition to their biodegradability and biocompatibility, nano-cellulosic materials present enhanced properties due to their high aspect

ratio (length/diameter) and specific surface area, high elastic modulus, high thermal resistance and these characteristics, combined with other peculiarities, make the research field in the area of nanocellulose growing towards an upward expansion [15,16].

The great diversity in the composition of natural fibers makes it necessary to adequately characterize them since the properties of their nano portions depend on the cellulosic source used and the applied extraction method [6,17,18]. Although these materials are basically structurally formed by cellulose (40 - 50%), hemicellulose (20 - 30%), and lignin (10 - 25%), their proportion and that of other components such as pectin, proteins, ash, and extracts affect crucially the conditions of pulping and the delignification step and the quality of the final product [19-22].

In this work, four Brazilian lignocellulosic materials, *Ceiba speciosa* (silk-floss), *Phormium tenax*, *Cortaderia selloana* (Pampas-grass) , and *Cocus nucifera*(coconut coir) were evaluated as raw-materials for nanocellulose production by mechanical and/or chemical treatments. Their chemical composition, extractives, and thermal properties were analyzed before attempts were made to produce nanocellulose samples. Different chemical and mechanical treatments were used during the production of these samples. It selected the samples that obtained the lowest lignin content for the production of cellulose nanofibers (CNF) for each of the fibers under study. They were characterized by scanning electron microscopy (SEM), transmission electron microscopy (TEM), thermogravimetric analysis (TGA) and x-ray diffraction analysis (XRD), except for the *Phormium tenax* nanocellulose, a perennial plant with promising mechanical properties, which is the most recent research object. [23]. The novelty of this work is the use of Brazilian fibers that are not commonly used to produce nanofibers and to attempt to use the least amount possible of chemicals so that the production is more environmentally friendly. We could produce silk-floss nanofibers without any chemical treatment. Still, it was important to evaluate the use of chemicals to compare the materials submitted to mechanical treatments.

2. Materials and Methods

2.1. Fibers characterization.

All fibers were dried at room temperature and prepared according to the TAPPI T257 standard. It was determined the moisture (TAPPI T264 CM-07), ashes (TAPPI T211 OM-02), and lignin insoluble in acid (TAPPI T13 WD-74) contents; NaOH solubility (TAPPI T212 OM-12); solubility in cold and hot water (TAPPI T207 CM-08), extractives in toluene/ethanol and total extractives (TAPPI T204 CM-07) [24-29]. For *Phormium tenax* the soluble lignin was determined by the Goldschimid method [30], and the cellulose and hemicellulose contents were determined by the NREL/TP510 standard [31]. All analyses were carried out in triplicate.

2.2. Chemical treatments applied to the fibers.

Table 1 presents all chemical treatments applied to the fibers.

Table 1. Chemical treatments.

Material	Chemical Treatment
Pampas-grass*	I) Mild $NaClO_2$ treatment: For every 10 g of pulp, 1.5 g of 80% sodium chlorite and 0,01 g/L of glacial acetic acid were used. The material was kept in a water bath at an 80°C for 1 hour. The process was repeated 3 times, and the fibers were rinsed with water before the next treatment.

Material	Chemical Treatment
	II) NaOH treatment I + Mild $NaClO_2$ treatment: aqueous NaOH solution 5% (w/w) for 1 hour under mechanical stirring at 70°C. After complete individualization, the solution was filtered, and the fibers were rinsed thoroughly with water.
	III) NaOH treatment II + H_2O_2 treatment: After the NaOH treatment I, the fibers were bleached with a NaOH aqueous solution at 4% (w/w) and hydrogen peroxide (H_2O_2) at 24% (w/w), in the proportion of 1:1, during 2 hours under constant mechanical agitation, at 50°C.
Silk-floss** 	I) NaOH treatment: fibers were treated using a 5% (w/w) NaOH solution at 70°C for 60 minutes and then were thoroughly washed with water [32].
	II) NaOH + mild $NaOCl_2$ treatment: using 1.5 g of sodium chlorite 80% for 1 h at 80 °C. Three delignification stages were performed, and in each interval, the pulp was washed with water [32].
	III) NaOH + H_2O_2 treatment) fibers were soaked in a 4% NaOH and 24% H_2O_2 solution (1:1) for 2h at 50°C. After extraction, fibers were thoroughly washed with water [32].
Coconut coir 	Cleaning of fibers twice at 50°C for 2h, NaOH 2% treatment also two times [13], and then mild $NaOCl_2$ treatment three times [33,34].
*Phormium tenax** 	I) The general conditions of the treatments were a solid-to-liquid ratio of 1:4 at 140°C for 1 hour, the ratio of solid to the liquid being 1:12. The liquid fraction was removed by filtration, and the solid fraction was rinsed until the dark liquor was eliminated. The first was made with fibers and water only.
	II) The second treatment had the same conditions as the first treatment, but here organosol was also used, adapted from [35] with ethanol:water ratio equal to 70:30, 46% ethanol on dry mass.

*dried fibers were cut with scissors and then milled in a knife mill
**In order to reduce the size of the fiber, the silk-floss fiber also underwent a mechanical pretreatment to the previously mentioned treatments. The silk floss was previously submitted to low temperature for 48 hours and then added to a knife mill with a 300 microns mesh at 12000 rpm.

2.3. Nanocellulose production.

According to the specific characteristics of each fiber and the chemical treatment applied, the one that resulted in the lowest lignin content to produce nanocellulose was chosen. For this, the fibers were dispersed in water (1.0 g.100ml-1) and mixed for 05 minutes at 1500 rpm. Immediately after homogenization, the suspensions were subjected to mechanical defibrillation in a colloidal stone mill (Supermass Colloider MKCA6-3, Masuko Sangyo Co. Ltd., Kawaguchi, Japan). The suspensions were recirculated 5 to 10 times (according to each fiber and previous results), with 0.1mm between stones and a rotation speed of 1500 rpm. At the end of the process, the nanofibril suspensions were collected and stored in closed bottles under refrigeration.

2.4. Nanocellulose characterization.

2.4.1. X-ray diffraction (XRD).

The crystallinity of the CNF samples was assessed by X-ray diffraction on a diffractometer (D8 Advance, Bruker Corp., Massachusetts, USA), operating at 40 kV, 20 mA and a source of Cu-Kα radiation with a wavelength 1.54060 Å. The dry CNF samples were scanned in 2θ ranges, ranging from 5 to 90º at a rate of 2θ.5s-1. The crystallinity index (CI) was estimated from peak heights (200) (2 × 22.5°) and the minimum intensity between peaks (110) and (200) (2θ between 16° and 22°) [36].

2.4.2. Thermogravimetric analysis (TGA).

The thermal degradation of the CNF samples was analyzed by thermogravimetric analysis (TGA Q500, TA Instruments, Delaware, USA) under nitrogen flow of 20 ml.min-1, with temperature ranging from 25 - 800ºC (10ºC.min-1) [37]. The initial degradation temperature was determined from the first inflection of the baseline curve of the mass loss derivative (ASTM-E2550).

2.4.3. Scanning electron microscopy (SEM).

The morphology of the cellulose nanoparticles was analyzed using a scanning electron microscope (VEGA3 LMU, Tescan, Brno, Czech Republic). The samples were fixed on metallic supports with the aid of copper tape, metalized with gold, and analyzed with an acceleration voltage of 10 kV. The images were evaluated using the Image J software (public domain).

2.4.4. Transmission electron microscopy (TEM).

The thickness of the cellulose nanofibrils could be assessed by transmission electron microscopy (JEM 1200EXII, Jeol Ltd., Tokyo, Japan). Dilutions in distilled water (5 ppm) of CNF 1% were dripped onto the surface of copper screens, and after the solvent had evaporated at room temperature, the samples were analyzed. As for SEM, images were evaluated using the Image J software (public domain).

3. Results and Discussion

3.1. The fibers characterization.

Table 2 presents the composition, extractives, and solubility of the fibers studied. Their detailed chemical characterization is extremely important to define the further treatments that must be applied to isolate cellulose, lignin, and nanocellulose.

The cold water removes tannins, gums, sugars, starch, and pigments. NaOH removes acidic substances that react to form salts, and the solubility content in NaOH gives an idea of the plant's resistance against fungi and other deteriorating agents. Extraction in ethanol/toluene removes greases, oils, waxes, insoluble compounds in ethyl ether, and some resins. The ashes represent the inorganic portion of the fibers (K, Mg, Si, and Ca). Leaf fibers such as Pampas-grass present high extractive levels from the plant defense system against insects. Sodium hydroxide is a cheap delignification protocol, and the high content of extractives for it demonstrates the successful application of this method aimed at lignin and extractives removal, and upon the lignin dissolves, the cellulose content increase.

Table 2. Composition, extractives, and solubility of fibers.

Content (%)	**Lignocellulosic raw material and standard deviation**			
	Pampas-grass	**Silk-floss**	**Coconut***	***Phormium tenax***
Humidity	14 ± 1	10.00 (±0.10)	-	10.99 ± 0.07
Ash	4.10 ± 0.03	0.85 – 0.92	1.82	4.11 ± 0.01
Solubility in cold water	16 ± 3	1.20 – 3.14	10.29	-
Solubility in hot water	19 ± 1	3.00 – 3.21	10.78	10.63 ± 0.10
Ethanol/toluene extractives	5 ± 1	2.10 – 7.03	7.79	10.90 ± 0.17
Ethanol extractives	0.9 ± 0.5	1.05 – 2.88	0.77	1.87 ± 0.17
NaOH extractives	43.1 ± 0.4	18.20 – 24.12	21.67	-

Content (%)	Lignocellulosic raw material and standard deviation			
	Pampas-grass	Silk-floss	Coconut*	*Phormium tenax*
Lignin	20.0 ± 0.8	14.94 – 16.20	7.48	15.02 ± 0.61
Total extractives	7.0 ± 0.5	2.86 – 4.05	12.78	23.40 ± 0.10

* Coconut fibers stored for 17 years and partially degraded.

The low lignin content of the coconut coir fibers, when compared with Abraham *et al*. [38] (±49.2%), could be related to the degradation during storage since the fibers at the present work were stored for 17 years in bags at ambient conditions (temperature and humidity). The long storage was part of research to evaluate the effect of storage time on the lignin content, focusing on more ecologically friendly chemical treatments and the use of fiber residues.

Concerning the morphological analysis of the fibers, it can be seen that the Pampas-grass (Figure 1a) presents longitudinal canals and defects in its surface, making it possible to observe lateral sections of the fibers, presenting a structure with the thick cell wall and large irregularly shaped lumen. As for silk-floss in nature, the fibers are smooth, long, and hollow (Figure 1b), with a cylindrical structure and without pores. This fiber has low lignin content (14 - 16%), which facilitates its mechanical defibrillation, and it is not necessary to use aggressive chemical treatments to get the nanocellulose obtained only with mechanical treatments. The amount of inorganic components (ash content of 0.85 - 0.95%) is the lowest compared to the other fibers. Silk floss presents a high hydrophobicity (97.6%), while *Phormium tenax* presents an exceptionally low hydrophobicity (12.0%), as most lignocellulosic materials. Hydrophobicity tests of these fibers were made according to the experiment of Ribeiro *et al*. [39]. Coconut fibers (Figure 1c) showed a quite rough surface, where layers of wax and extractives cover it; the same aspect was observed by Muensri *et al*. [40]. The globular protrusions disposed at the surface are tyloses, fatty deposits that appear in vegetable cells when the plant is exposed to stress conditions such as low air humidity or bacterial/fungal infestations [41]. Figure 1d shows the morphology of *Phormium tenax* fibers, where at the smaller magnification (x 200), it is possible to see the shredded cell walls and the group of fibers responsible for forming the fiber bundle. At the higher magnification (x 2000), a similar characteristic to that found by Guen *et al*. [42] can be seen because individual, linear fibers with smooth curvatures and small points of apparent roughness appear, which is due to the leaf fibers being narrower within an order of magnitude.

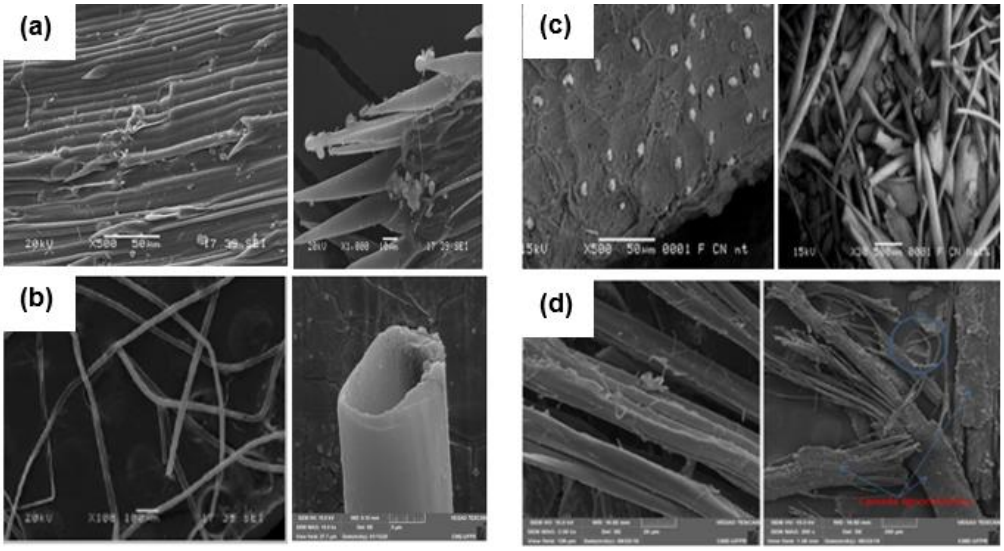

Figure 1. Fiber SEM of (**a**) Pampas-grass; (**b**) Silk-floss; (**c**) Coconut; (**d**) *Phormium tenax.*

Figure 2 and 3 show, respectively, the XRD and TGA of 3 fibers. The high thermal stability of the Pampas grass (Figure 3a) grass can be related to the high 30.5% crystallinity index (Figure 2a) and high lignin content of the fiber. The nanocellulose obtained by treatment II showed a crystallinity index of 30.9%. The crystallinity index of silk floss (Figure 2b) was 25.4% and is low due to the amorphous components on its surface.

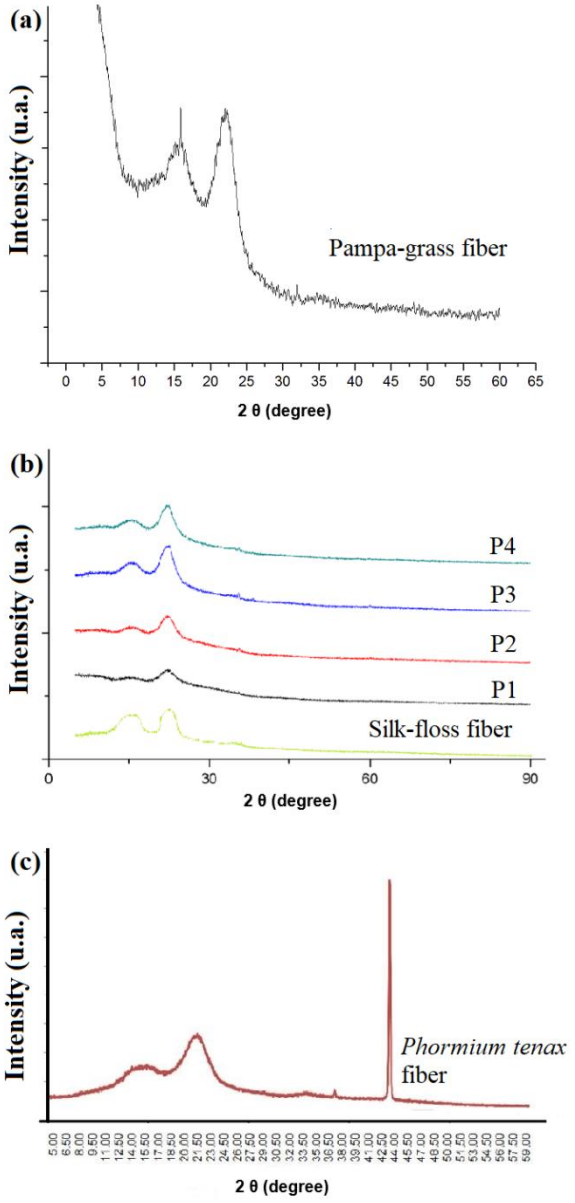

Figure 2. XRD of **(a)** Pampas-grass*;* **(b)** Silk-floss; (**c**) *Phormium tenax.*

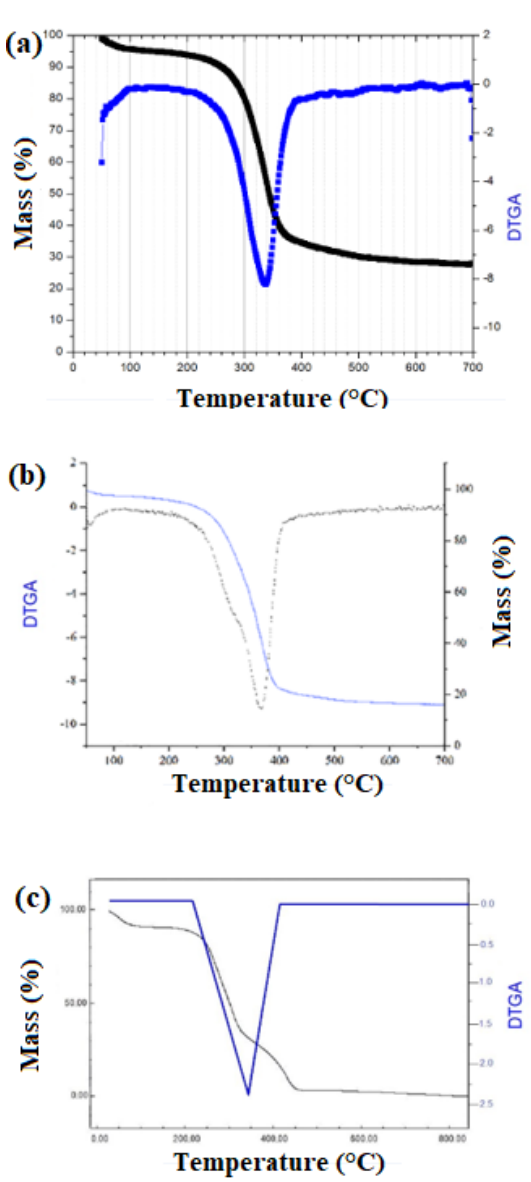

Figure 3. TGA of **(a)** Pampas-grass*;* **(b)** Silk-floss; **(c)** *Phormium tenax.*

It can be seen that with the selected treatment (treatment II) and varying the number of fibers passing in the colloid mill, more crystalline regions of the fiber are exposed, and the crystalline index increases. The moisture content in the vegetable fibers is influenced by climatic conditions (loss of water in TGA). The silk-floss fiber behaves well at high temperatures, showing thermal stability up to 240 °C, whereas degradation of the cellulose occurs in the temperature range of 360 to 500 °C (Figure 3b). The crystallinity index of *Phormium tenax* was 36.7% (Figure 2c). This fiber shows a loss of mass in the range between 38 to 130 ºC that corresponds to the vaporization of water. It can be seen in Figure 2c that the

most significant loss of mass occurs in the range of 200 to 325 °C, where it represents around 55 to 56%. A third degradation curve appears from 325 to 450°C with a mass loss of approximately 33%. After 450°C, the remaining mass accounts for the ash content and is between 2 to 4%.

3.2. Nanocellulose characterization.

Table 3 shows the amount of lignin for each chemical treatment performed, as described in table 1.

Table 3. Content of lignin after chemical treatments is described in Table 1.

Treatment	Lignin (%)			
	Pampas-grass	**Silk-floss**	**Coconut**	***Phormium tenax***
I	3.3	3.22	0,7	9.5
II	2.4	1.56	-	6.1
III	6.6	9.96	-	-

The NaOH treatment I + Mild $NaClO_2$ treatment used for Pampas-grass fibers reduced the lignin content from 20% to 2.4%, more effective than the one that used sodium hydroxide + hydrogen peroxide (6.6%) or only sodium chlorite (3.3%). For silk floss, using the same chemicals effectively reduced the lignin content (1.56%) because the processes using these chemicals attack the hydrogen bonds in the fiber structure, removing a significant amount of lignin, waxes, greases, and hemicellulose in the fiber [43].

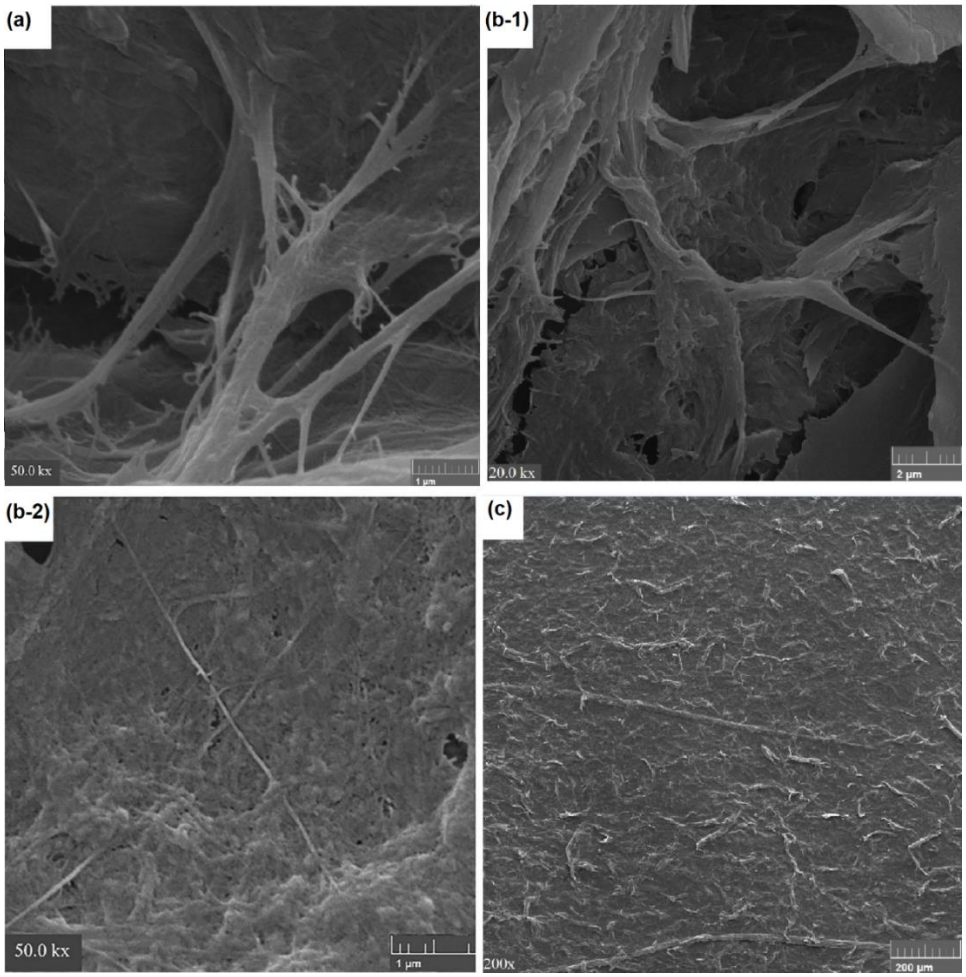

Figure 4. Nanocellulose SEM of (**a**) Pampas-grass with chemical treatment II; (**b-1**) Silk-floss mechanical treatment only and (**b-2**) Silk-floss with chemical treatment II; (**c**) Coconut coir with the chemical treatment I.

The use of H_2O_2 is suitable for the extraction of cellulose that is hardly decomposed under this condition. However, since the silk-floss lignin content is low, it is not essential that strong chemical treatments are used. Although sodium hydroxide may be used to remove lignin, waxes, and grease, the delignification process may be excluded if cold milling is used before submitting the material to mechanical defibrillation. In this case, it can be seen that the fibers can interact completely with the organic solvent (ethanol) or a mixture of an organic solvent/water. It is possible to produce nanofibers without the need for chemical treatment for delignification. For *Phormium tenax* it is possible to see the reduction of lignin content with the use of an organic solvent (ethanol) compared to the treatment performed with water only. However, its characterization is still being carried out.

Figure 4 shows SEM photos of the fibers from the treatments that resulted in the lowest lignin content for each fiber studied. In the case of silk-floss, it is also shown photos of nanocellulose obtained only through mechanical treatment of the fiber (milling of fibers at low temperatures, 300 μm screen) (Figure 4b-1) and use of ethanol as solvent (Figure 4b-2). The main difference between the non-chemically treated (only mechanically treated) and chemically treated silk-floss nanocellulose is the nanofibers form after the process: from the first case, it was obtained a hydrophilic film, from the second case it was obtained nanocellulose in the form of powder after drying. This powder is partially hydrophobic and facilitates the preparation of hydrophobic nanocomposites.

Figure 5 shows the TEM micrographs of the Pampas grass, silk floss, and coir. It was possible to observe that the different delignification processes did not affect the mechanical defibrillation process, possibly the production of nanostructures, regardless of the treatment used for removing lignin in each of the fibers. However, the amount of nanostructures varies and can be visually observed according to the fiber delignification process. For Pampas-grass, for example, an average of 20 nm of nanostructure was obtained, as shown in Figure 5a. On the other hand, the CNF of the silk floss with treatment, shown in Figure 5b, shows a tangle of fibrils, making it difficult to read the length of these cellulose nanofibrils. This is possibly due to the formation of interfibrillar hydrogen bonds, which is commonly observed in nanocelluloses obtained from other sources such as bamboo, rice straw, corn straw, and others [32]. The coir also shows clustered and overlapping fibrils, making it difficult to accurately measure the obtained nanostructure, as seen in Figure 5c.

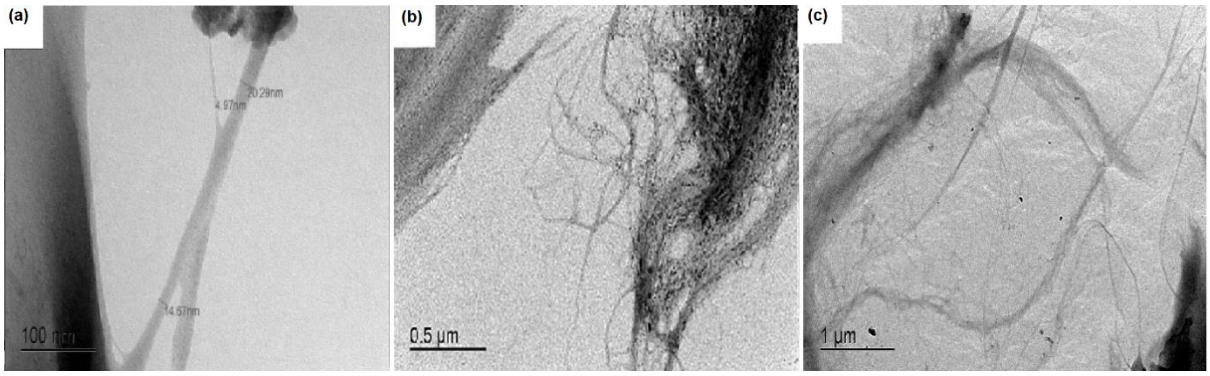

Figure 5. Nanocellulose TEM of (**a**) Pampas-grass*;* (**b**) Silk-floss; (**c**) Coconut coir.

4. Conclusions

Lignin, $C_9H_{10}O_2(OCH_3)n$, the major noncarbohydrate component of lignocellulose, may be partially removed by chemical treatments. Various nanocellulose extraction processes

can be used, although harmful chemicals and high-energy treatments for nanocellulose processing must be avoided. Low-cost and environmentally friendly treatments promote the development of cleaner technology for nanocellulose production. By comparing different raw materials, it is possible to choose the ones with the best potential for producing nanocellulose for different applications. Although the lignin content of fibers is an important factor in nanocellulose production, developing mechanical rather than chemical treatments may be enough to produce nanofibers. In the case of silk floss, it was possible to produce nanofibers without any chemical treatment. On the other hand, using coconut fibers stored for a long time and partially degraded resulted in the need for a less severe chemical treatment since the content of lignin was much lower than that of fresh coconut fibers. Hydrophobicity is an interesting test to evaluate the potential use of plants for nanofiber production because CNF produced may reinforce polar or apolar polymers without the need for chemical modification.

Funding

This study was supported by the National Council for Scientific and Technological Development (CNPq), the Coordination for the Improvement of Higher Education Personnel (CAPES), and for the National Council of Science and Technology (Conacyt), which awarded fellowships to authors.

Acknowledgments

The authors thank the Microscopy Laboratory of the Federal University of Paraná (UFPR).

Conflicts of Interest

The authors declare no conflict of interest.

References

1. Pires, J.R.A.; Souza, V.G.L.; Fernando, A.L. Valorization of energy crops as a source for nanocellulose production – Current knowledge and future prospects. *Ind Crops Prod* **2019**, *140*, 1-17, https://doi.org/10.1016/j.indcrop.2019.111642.
2. Moshood, T.D.; Nawanir, G.; Mahmud, F.; Mohamad, F.; Ahmad, M.H.; AbdulGhani, A. Sustainability of biodegradable plastics: new problem or solution to solve the global plastic pollution? *Curr Rea Gr Sust Chem* **2022**, *5*, 1-18, https://doi.org/10.1016/j.crgsc.2022.100273.
3. Singh, N.; Ogunseitan, O.A.; Wong, M.H.; Tang, Y. Sustainable materials alternative to petrochemical plastics pollution: a review analysis. *Sust Hor* **2022**, *2*, 1-16, https://doi.org/10.1016/j.horiz.2022.100016.
4. Rosenboom, J.G.; Langer, R., Traverso, G. Bioplastics for a circular economy. *Nat Rev Mat* **2022**, *7*, 117-137, https://doi.org/10.1038/s41578-021-00407-8.
5. Mohanty, A.K.; Wu, F.; Mincheva, R.; Hakkarainen, M.; Raquez, J.M.; Mielewski, D.F.; Narayan, R.; Netravali, A.N.; Misra, M. Sustainable polymers. *Nat Rev Meth Prim* **2022**, *2*, https://doi.org/10.1038/s43586-022-00124-8.
6. Reshmy, R.; Thomas, D.; Philip, E.; Paul, S.A.; Madhavan, A.; Sindhu, R.; Sirohi, R.; Varjani, S.; Pugazhendhi, A.; Pandey, A.; Binod, P. Bioplastic production from renewable lignocellulosic feedstocks: a review. *Rev Env Sci Biotech* **2021**, *20*, 167-187, https://doi.org/10.1007/s11157-021-09565-1.
7. Zergane, H.; Abdi, S.; Xu, H.; Hemming, J.; Wang, X.; Willför, S.; Habibi, Y. Ampelodesmos mauritanicus a new sustainable source for nanocellulose substrates. *Ind Crops Prod* **2020**, *144*, 1-9, https://doi.org/10.1016/j.indcrop.2019.112044.
8. Marakana, P.G.; Dey, A.; Saini, B. Isolation of nanocellulose from lignocellulosic biomass: Synthesis, characterization, modification, and potential applications. *Jour of Envir Chem Eng* **2021**, *9*, 106606, https://doi.org/10.1016/j.jece.2021.106606.

9. Baranwal, J.; Barse, B.; Fais, A.; Delogu, G.L.; Kumar, A. Biopolymer: a sustainable material for food and medical applications. *Polym* **2022**, *14*, 1-22, https://doi.org/10.3390/polym14050983.
10. Ahmad Khorairi, A.N.S.; Sofian-Seng, N.S.; Othaman, R.; Abdul Rahman, H.; Mohd Razali, N.S.; Lim, S.J.; Wan Mustapha, W. A. A review on agro-industrial waste as cellulose and nanocellulose source and their potentials in food applications. *Food Rev Inter* **2021**, 1-26, https://doi.org/10.1080/87559129.2021.1926478.
11. Oberti, I.; Paciello, A. Bioplastics as a substitute for plastic in construction industry. *Encyc* **2022**, 2, 1408-1420, https://doi.org/10.3390/encyclopedia2030095.
12. Klemm, D.; Cranston, E.D.; Fischer, D.; Gama, M.; Kedzior, S.A.; Kralisch, D.; Kramer, F.; Kondo, T.; Lidnström, T.; Nietzsche, S.; Welcke, K.P.; Rauchfuß, F. Nanocellulose as a natural source for groundbreaking applications in materials science: Today's state. *Mat today* **2018**, *21*, 720-748, https://doi.org/10.1016/j.mattod.2018.02.001.
13. Sharma, C.; Sharma, S.N.; Srivastava, R. Analysis of cellulose extracted from waste products. *Coll and Poly Scien*, **2022**, 1-10, http://doi.org/10.1007/s00396-022-05005-w.
14. Picot-Allain, M.C.N.; Emmambux, M.N. Isolation, characterization, and application of nanocellulose from agro-industrial by-products: A review. *Food Rev Inter* **2021**, 1-2, https://doi.org/10.1080/87559129.2021.1928689.
15. Sun, D.; Onyianta, A.J.; O'Rourke, D.; Perrin, G.; Popescu, C.M.; Saw, L.H.; Cai, Z.; Dorris, M. A process for deriving high quality cellulose nanofibrils from water hyacinth invasive species. *Cellul* **2020**, *27*, 3727-3740, https://doi.org/10.1007/s10570-020-03038-4.
16. Qasim, U.; Osman, A.I.; Al-Muhtaseb, A.H.; Farrell, C.; Al-Abri, M.; Ali, M.; Vo, D.V.N.; Jamil, F.; Rooney, D.W. Renewable cellulosic nanocomposites for food packaging to avoid fossil fuel plastic pollution: a review. *Env Chem Lett* **2021**, *19*, 613-641, https://doi.org/10.1007/s10311-020-01090-x.
17. Mishra, S.; Kharkar, P.S.; Pethe, A.M. Biomass and waste materials as potential sources of nanocrystalline cellulose: comparative review of preparation methods (2016 – till date). *Carb Pol* **2019**, *207*, 418-427, https://doi.org/10.1016/j.carbpol.2018.12.004.
18. Jonoobi, M.; Oladi, R.; Davoudpour, Y.; Oksman, K.; Dufresne, A.; Hamzeh, Y.; Davoodi, R. Different preparation methods and properties of nanostructured cellulose from various natural resources and residues: a review. *Cellul* **2015**, *22* , 935–69, https://doi.org/10.1007/s10570-015-0551-0.
19. Nascimento, S.A.; Rezende, C.A. Combined approaches to obtain cellulose nanocrystals, nanofibrils and fermentable sugars from elephant grass. *Carb Pol* **2018**, *180*, 38–45, https://doi.org/10.1016/j.carbpol.2017.09.099.
20. Yu, S.; Wang, L.; Zhang, Y.; Zhou, H. Sustainable carbon materials from the pyrolysis of lignocellulosic biomass. *Mat Tod Sust*, **2022**,100209, https://doi.org/10.1016/j.mtsust.2022.100209.
21. Wang, X.; Zhang, G.; Dang, Y. Enhanced Extraction of Flavonoids from Licorice Residues by Solid-State Mixed Fermentation. *Was and Bio Val* **2022**, 1-13, https://doi.org/10.1007/s12649-022-01803-z.
22. Aggarwal, N.K.; Kumar, N.; Mittal, M. A Feasible Approach for Bioethanol Production Using Conventional and New Feedstocks. *Bio Prod* **2022**, 47-63, https://doi.org/10.1007/978-3-031-05091-6_4.
23. Rosa, I.M.; Iannoni, A.; Kenny, J.M.; Puglia, D.; Santulli, C.; Sarasini, F.; Terenzi, A. Poly(lactic acid)/Phormium tenax composites: morphology and thermo-mechanical behavior. *Pol Comp* **2011**, 1362–1368, https://doi.org/10.1002/pc.
24. Technical Association of the Pulp and Paper Industry TAPPI T264 CM-07: Preparation of Wood for Chemical Analysis, Atlanta (USA) **2007**, https://imisrise.tappi.org/TAPPI/Products/01/T/0104T264.aspx.
25. Technical Association of the Pulp and Paper Industry TAPPI T211 OM-02: Ash in wood, pulp, paper and paperboard: combustion at 525 °C, Atlanta (USA) **2007**, https://imisrise.tappi.org/TAPPI/Products/01/T/0104T211.aspx.
26. Technical Association of the Pulp and Paper Industry TAPPI T222 om-21: Acid-insoluble lignin in wood and pulp, Atlanta (USA) **2009.** https://imisrise.tappi.org/TAPPI/Products/01/T/0104T222.aspx
27. Technical Association of the Pulp and Paper Industry TAPPI T212 OM-12: One Percent Sodium Hydroxide Solunility of Wood and Pulp, Atlanta (USA) **2012**, https://tappi.micronexx.com/CD/TESTMETHODS/T212.pdf.
28. Technical Association of the Pulp and Paper Industry TAPPI T207 CM-08: Water Solubility of Wood and Pulp, Atlanta (USA) **2008**, https://imisrise.tappi.org/TAPPI/Products/01/T/0104T207.aspx.
29. Technical Association of the Pulp and Paper Industry TAPPI T204 CM-07: Solvent Extractives of Wood and Pulp, Atlanta (USA) **2007,** https://www.tappi.org/content/sarg/t204.pdf.
30. Goldschimid, O.; Sarkanen, K.V.; Ludwwig, C.H.; John Wiley. Ultraviolet spectra. In Lignins: occurrence,

formation, structure and reactions. *New York* **1971**, 241–266, https://doi.org/10.1002/pol.1972.110100315.
31. Sluiter, C.S.A.; Hames, B.; Ruiz, R.; Slui, J.; Templeton, D. Determination of Structural Carbohydrates and Lignin in Biomass. Laboratory Analytical Procedure (LAP). *Tech Rep NREL/TP-510-42618* **2008**, 1–15, https://www.nrel.gov/docs/gen/fy13/42618.pdf.
32. Leal, M.R.; Flores-Sahagun, T.H.S.; Franco, T.S.; Muniz, G.I.B. Ceiba Speciosa St. Hill Fruit Fiber as a potential source for nanocellulose production and reinforcement of polyvinyl acetate composites. *Pol Comp* **2020**, *42*, 1-15, https://doi.org/10.1002/pc.25833.
33. Rosa, M.F., Medeiros, E.S.; Malmonge, J.A.; Gregorski, K.S.; Wood, D.F.; Mattoso, L.H.C.; Glenn, G.; Orts, W.J.; Imam, S.H. Cellulose nanowhiskers from coconut husk fibers: effect of preparation conditions on their thermal and morphological behavior. *Carb Pol* **2010**, *81*, 83–92, https://doi.org/10.1016/j.carbpol.2010.01.059.
34. Wise, L.E.; Murphy, M.; Adieco, A.A. A chlorite holocellulose, its fractionation and bearing on summative wood analysis and studies on the hemicelluloses. *Pap Trade J* **1946**, *122*, 35-43, https://www.semanticscholar.org/paper/A-CHLORITE-HOLOCELLULOSE%2C-ITS-FRACTIONATION-AND-ON-Wise-Murphy/53f10e1d55261efe9fe84a6d2317b4d901d95274.
35. Robles, E.; Rodríguez, J.F.; Barbosa, A.M.; Gordobil, O.; Carreño, N.L.V.; Labidi, J. Production of Cellulose Nanoparticles from Blue Agave Waste Treated with Environmentally Friendly Processes. *Carb Pol* **2018**, *183*, 294-302, https://doi.org/10.1016/j.carbpol.2018.01.015.
36. Segal, L.; Creely, J.J.; Martin, A.E.; Conrad, C.M. An empirical method for estimating the degree of crystallinity of native cellulose using the X-ray diffractometer. *Text Res J* **1959**, *29*, 786-794, https://doi.org/10.1177/004051755902901003.
37. Cao, Y.; Jiang, Y.; Song, Y.; Cao, S.; Miao, M.; Feng, S.; Fang, J.; Shi, L. Combined bleaching and hydrolysis for isolation of cellulose nanofibrils from waste sackcloth. *Carb Pol* **2015**, 1-19, https://doi.org/10.1016/j.carbpol.2015.05.063.
38. Abraham, E.; Deepa, B.; Pothen, L.A.; Cintil, J.; Thomas, S.; John, M.J.; Anandjiwala, R.; Narine, S.S. Environmentally friendly method for the extraction of coir fibre and isolation of nanofibre. *Carb Pol* **2013**, *92*, 1477-83, https://doi.org/10.1016/j.carbpol.2012.10.056.
39. Ribeiro, T.H.; Rubio, J.; Smith, R.W. A dried hydrophobic aquaphyte as an oil filter for oil/water emulsions. *Spill Sci Tech Bull* **2003**, *8*, 483-89, https://doi.org/10.1016/S1353-2561(03)00130-0.
40. Muensri, P.; Kunanopparat, T.; Menut, P.; Siriwattanayotin, S. Effect of lignin removal on the properties of coconut coir fiber/wheat gluten biocomposite. *Comp Part A: Ap Sci Man* **2011**, *42*, 173-79, https://doi.org/10.1016/j.compositesa.2010.11.002.
41. Leão, R. M.; Luz, S.M.; Araujo, J.A.; Novack, K. surface treatment of coconut fiber and its application in composite materials for reinforcement of polypropylene. *J Nat Fib* **2015**, *12*, 574-86, https://doi.org/10.1080/15440478.2014.984048.
42. Guen, M.J.L.; Newman, R.H. Pulped Phormium Tenax leaf fibres as reinforcement for epoxy composites. *Comp Part A: Ap Sci Man* **2007**, *38*, 2109-15, https://doi.org/10.1016/J.COMPOSITESA.2007.07.001.
43. Bartos, A.; Anggono, J.; Farkas, A.E.; Kun, D.; Soetaredjo, F.E.; Móczó, J.; Purwaningsih, A.H.; Pukánszky, B. Alkali treatment of lignocellulosic fibers extracted from sugarcane bagasse: composition, structure, properties. *Pol Test* **2020**, *88*, 1-8, https://doi.org/10.1016/j.polymertesting.2020.106549.