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Extraction and Physico-Chemical Characterization of Pineapple Crown Leaf Fibers (PCLF)

Vivek Johny [1], Ajith Kuriakose Mani [1], Sivasubramanian Palanisamy [2], Visakh Kunnathuparambil Rajan [1], Murugesan Palaniappan [3] and Carlo Santulli [4,*]

[1] Department of Mechanical Engineering, Saintgits College of Engineering, Pathamuttom, Kottayam 686532, Kerala, India
[2] Department of Mechanical Engineering, Dilkap Research Institute of Engineering and Management Studies, Neral, Karjat 410101, Maharashtra, India
[3] Department of Mechanical Engineering, College of Engineering, Imam Mohammed Ibn Saud Islamic University, Riyadh 11432, Saudi Arabia
[4] School of Science and Technology, Università degli Studi di Camerino, 62032 Camerino, Italy
* Correspondence: carlo.santulli@unicam.it; Tel.: +39-380-6522232

Abstract: Apart from the widely discussed pineapple leaf fibers, normally referred to as PALF, fibers from other parts of the plant also exist, particularly those in the fruit crown, which are known as pineapple crown leaf fibers (PCLF). In this work, PCLF were characterized using thermogravimetric analysis (TGA), Fourier transform IR spectroscopy (FTIR), X-ray diffraction (XRD), scanning electron microscopy (SEM), and atomic force microscopy (AFM). The results indicated that the properties of PCLF do not greatly differ from those observed for PALF. In particular, a cellulose content of over 67% was observed, with approximately 76% crystallinity. The main degradation phenomena of the fibers took place between 230 and 380 °C, peaking at 324 °C, which is in line with observations in other fibers which have similar cellulose and crystalline contents. There was 13.4% residue at 680 °C. Bare mechanical retting of PCLF, although not allowing a full and thorough degumming, which would only be achieved through more aggressive chemical treatment, enabled aspect ratios of over 10^3 to be obtained. This indicates some potential for their application as short fibers in composites. In this respect, the considerable roughness of PCLF when compared to other leaf-extracted fibers, and in particular when compared to PALF, could suggest an ability to obtain a sufficiently sound fiber–matrix interface.

Keywords: pineapple waste; degradation temperature; cellulose crystallinity; microscopic morphology; roughness

Citation: Johny, V.; Kuriakose Mani, A.; Palanisamy, S.; Rajan, V.K.; Palaniappan, M.; Santulli, C. Extraction and Physico-Chemical Characterization of Pineapple Crown Leaf Fibers (PCLF). *Fibers* **2023**, *11*, 5. https://doi.org/10.3390/fib11010005

Academic Editors: Damien Soulat

Received: 11 October 2022
Revised: 17 December 2022
Accepted: 28 December 2022
Published: 6 January 2023

1. Introduction

When using lignocellulosic fibers extracted from plants, large variability in the resulting properties is often encountered. This might depend, other than on the species, on a number of other factors, such as the cultivar, the age of the plant, and the mode of extraction of the fiber [1]. When aiming at developing an engineering application for the fibers, an obvious limitation is that the interaction of these factors cannot normally be fully evaluated. This is a consequence of the fact that the available information on the plants from which the fibers are to be obtained is seldom sufficiently complete. This issue is especially relevant when, as has frequently occurred in the last few years, the fibers are a by-product or a waste from certain sectors, such as agriculture, forest management, food production, or biofuel synthesis. In these instances, evaluating properties such as the thermal stability of the fibers, which is essential for the production of composites, may produce a non-negligible issue [2].

With the purpose of addressing the aforementioned issue of the variability of lig-

nocellulosic material performance, in those cases in which the fibers are extracted from different parts of the plant, a recent trend involves attempting to compare the properties of these different materials. This is the case for a number of production systems. Examples are hemp, where fibers and shives are available [3]; banana, including fibers from the leaf [4] and the fruit peel [5,6]; oil palm, which, as well as leaf fibers, has empty fruit bunch fibers [7,8]; coconut (coir fibers) [9]; bamboo [10]; olive oil (stones and seeds, pomace, and resins) [11], rattan (wood and leaves) [12,13], and pineapple, to which the present work is dedicated [14].

Regarding the latter plant (*Ananas comosa*), there are a number of reviews relating the use of pineapple leaf fibers (PALF) in composites, which sometimes deal with specific aspects crucial for composite production, such as PALF extraction and treatment, and their mechanical and thermal properties. This indicates a considerable interest, which has been continuously increasing over the last few years [15–19].

In view of the productive use of the complete plant, an obvious and abundant waste material from pineapple production is the crown of leaves that constitute the top of the fruit, which is a by-product of fruit processing. The fibers extracted from the crown are referred to as pineapple crown leaf fibers (PCLF). Data available on PCLF are mainly relevant for their possible use as a source of cellulose [20]. Therefore, clarifying their adaptability to pulping using different chemical solutions [21] or fine-tuning a method for the extraction of nano-crystalline cellulose (NCC) via hydrolysis in sulfuric acid [22] has been suggested. However, another possibility is to position these fibers as competitors to PALF, especially through the ability to extract them in a less destructive way, and also for the reason that the material available for the production of PCLF may be more abundant compared to PALF, which is originated from plant pruning or possibly from the end of the productive life of the plant. Despite this, studies on PCLF regarding their prospective introduction into natural fiber composites, and therefore as possible "second choice" fibers from pineapple plant by-products for that purpose, are virtually non-existent. The present study proposes to cover this gap for by fully characterizing PCLF after extraction, and by comparing their properties against other fibers such as PALF and other leaf-extracted fibers.

A number of studies exist on the characterization of these fibers once extracted from the plant, which enables an assessment of the viability of their potential uses (e.g., in composites). These include studies on fibers from abaca (*Musa textilis*) [23], banana (*Musa indica*) [24], king pineapple (*Agave cantala*) [25], caroa (*Neoglaziovia variegata*) [26], phormium (*Phormium tenax*) [27], sansevieria (*Sansevieria trifasciata*) [28], and curaua (*Ananas erectifolius*) [29]. Moreover, some species of agaves are currently being widely explored for use in composites, including henequen (*Agave fourcroydes)* [30], sisal *(Agave sisalana)* [31], and blue agave (*Agave tequilana*), a waste from liqueur production [32].

2. Materials and Methods

2.1. Extraction of Fiber

Pineapple crown leaf fibers (PCLF) from the Queen variety grown in India, were extracted using water in a mechanical retting process. Initially, each pineapple fruit was peeled from the cluster of crown leaves (Figure 1a). These leaves were then put into a large basin, which was subsequently filled with fresh water until the leaves were completely submerged, as shown in Figure 1b. Following this, the top of the basin was completely covered with a metal sheet in order to exclude direct sunlight and rain. The basin was opened every five days (Figure 1c) and the water was replaced. The total duration of the retting process was 20 days, in order to simulate, as closely as possible, the type of field retting performed on more diffuse fibers such as hemp [33] (Figure 1d). At the end, the water was removed (Figure 1e).

Figure 1. (**a**) Cluster of pineapple crown leaves (PCLs) removed from pineapple head; (**b**) PCLs immersed into basin; (**c**) water replacement after 5 days; (**d**) PCLs after 20 days; (**e**) PCLs after 20 days with water removed, ready for fiber extraction.

2.2. Scraping and Retting Process of Pineapple Leaves

After 20 days, the leaves were removed from the basin and the fibers were extracted carefully from the decayed leaves (Figure 2a). Approximately 10 fibers were acquired from each leaf. A decorticating process was performed manually using brushes and aluminum plates. The fibers were collected and rinsed with distilled water and the adhered dirt was removed. The cleaned fibers were then sun-dried until they were separated (Figure 2b).

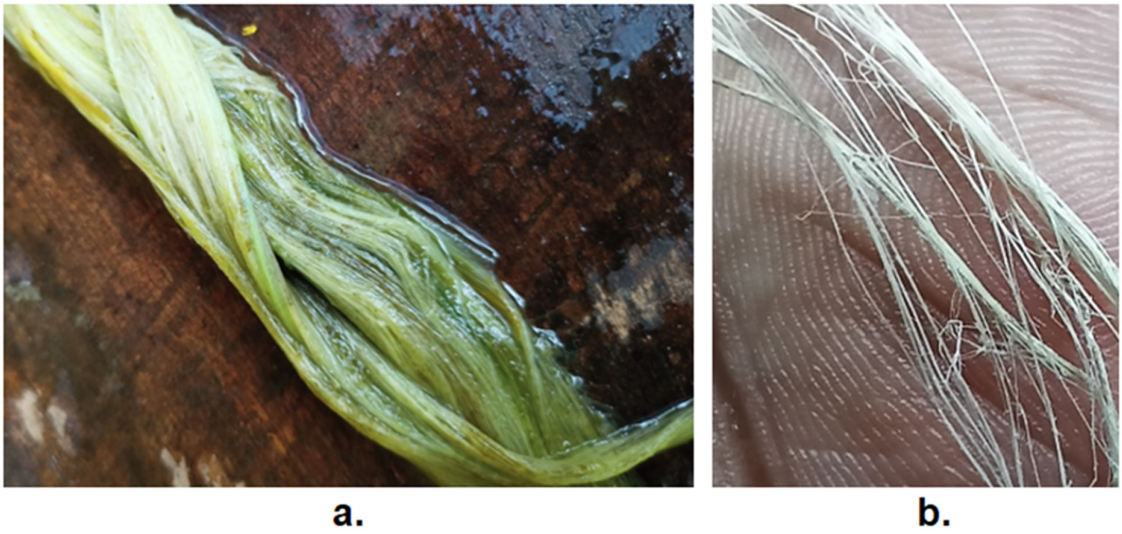

Figure 2. (**a**)Manually extracted fibers; (**b**)Extracted fibers after drying at ambient temperature.

Thepineapple leaf scraping process (Figure 2) was performed manually using a smooth wire brush, as suggested in [34]. During this process, the decayed debris surrounding the leaves was removed. The healthier leaves which were to be scraped were cleaned using fresh water and kept together until after the full removal of debris. The scratched pineapple crown leaf bundles were then immersed into a retting tank containing a 1/20 substrate/liquor ratio. A 0.5% urea solution was used to speed up the separation process. Soaking time was one hour. The leaves were then checked to ascertain whether the fibers were loosened and could be separated using the fingertips. The extracted fibers were then cleaned in fresh filtered water (drinking quality) and dried at ambient temperature (25 ± 2 °C).

2.3. Scanning Electron Microscopy (SEM)

A scanning electron microscope (SEM) model Quanta 200 (FEI, Hillsboro, Oregon, United States) was employed, using an accelerating voltage of 20 kV, 5 nm resolution, and a nominal vacuum level of 1.5×10^{-3} Pa. To prepare for the observation, three fiber specimens were covered in a thin gold layer to prevent the accumulation of electrical charges during the examination. The purpose of the study carried out using SEM was to observe the surface morphology of the extracted fibers and to evaluate their similarity to other natural fibers; in particular, to various fibers obtained from plant leaves, and more specifically, to PALF.

2.4. Thermogravimetric Analysis (TGA)

Thermogravimetric analysis (TGA) was used for the measurement of the thermal stability and temperature degradation intervals of the fibers using a temperature scanning procedure. An STA 449 F3 Jupiter (NETZSCH GmbH & Co. Holding KG, Selb, Germany) instrument was used for analysis, which was able to accurately record the

sample weight during heating between ambient temperature and 1550 °C. Approximately 5.3 mg of PCLF was heated in an aluminum oxide (Al_2O_3) crucible; and the temperature was increased from 40 to 700 °C at 10 °C per minute. Nitrogen (N_2) was delivered to the heating system at a flow rate of 30 mL/min to preserve an inert environment.

2.5. X-ray Diffraction (XRD)

XRD analysis was carried out to measure the crystallinity of the cellulose fraction of the fibers using a powder X-ray diffractometer X-Pert3 PRO by Panalytical (Malvern, UK) with copper (Cu) Kα1 energy equal to 8.04 keV and 0.039° resolution. A voltage of 40 kV was used, while the intensity of the current was 15 mA. The extracted fibers were pressed into 10 mm diameter pellets prior to performing the X-ray measurements.

2.6. Atomic Force Microscopy (AFM)

The surface roughness profile (Ra) was obtained and analyzed using a piezo-electric J-type scanner fitted with curving tips from 10 to 20 nm, developed from a Nanoscope IIIA (Veeco Co., Ltd., Shanghai, China) atomic force microscope (AFM). This allowed images to be obtained with an accuracy to the nanometer level.

2.7. Single Fiber Tensile Tests (SFTT)

For the purpose of load application, the aligned fibers were bonded to an adapted paper frame according to ISO 11566, The clamping mechanism of a tabletop ETM-B Series testing machine equipped with a head allowing a maximum force of 10 N was used (Shenzhen Wance Testing Machine Co., Ltd., Shenzhen, China). Tensile tests were carried out in displacement control mode by applying a load velocity equal to 1 mm/minute on a total of twenty PCL fibers with a total length of 90 (±5) mm and grip length of 30 mm. From these, seven valid results were obtained; three of the fibers were unable to take substantial load, fibrillating after gripping and pulling in the tensile machine.

3. Results and Discussion

The chemical composition of PCLF is reported and compared with that of other fibers reported in literature in Table 1. We limited the investigation to leaf fibers with a cellulose content exceeding 50%. Preference has been given to published data displaying the most completeness and consistency.

Table 1. Chemical composition of PCLF compared with other fibers of high cellulose content.

Fiber	Cellulose (%)	Hemicellulose (%)	Lignin (%)	Wax (%)	Pectin (%)	Ash (%)	Ref.
PCLF	67.3	16.9	7.4	3.8	1.3	0.8	-
Abaca	58.3	18.3	8.3	8.3	2.8	-	[35]
Banana peduncle	73.2	10.8	15.3	0.2	-	2.6	[36]
Sansevieria cylindrica	79.7	10.1	3.8	0.1	-	-	[37]
Curaua	73.6	9.9	7.5	-	-	0.9	[29]
Agave americana	68.4	15.7	4.9	0.3	-	-	[38]
Sisal	73.4	10	8	1.1		1.5	[39]
PALF	68.5	18.8	6	3.2	1.1	0.9	[40]

In order to provide evidence that PCLF can be used in composites, it is important to verify whether a sufficient aspect ratio (length/diameter) can be obtained after extraction. This depends on the maximum possible length extracted, which, in leaf fibers, is patently linked to the form of the cells. In principle, the length and diameter of the leaf fibers depend on the length and diameter of the cells. For example, for abaca fibers, depending on the species, the ratio between cell length and diameter might vary between approxi-

mately 80 and 140, which leads to a high fiber aspect ratio, as the technical fiber extracted is composed of a very variable number of cell units adhered together [41]. The average aspect ratios and densities of a number of similar fibers are compared to PCLF in Table 2. Aspect ratios around 10^3 were achieved. While this does not equal that obtained from PALF or the other leaf fibers, it remains sufficiently high to allow the possibility of twisting of some fibers into bundles.

Table 2. Average physical properties of PCLF compared to other high-cellulose fibers.

Fiber	Diameter (μm)	Length (mm)	Aspect Ratio (×10³)	Density (ϱ) (kg/m³)	Ref.
PCLF	108	155	1.43	1273	This study
PALF	59.7	300.5	5.03	1440 [42]	[43]
Sansevieria trifasciata	120	1090	9.83	1415	[44]
Curaua	65	1250	19.23	1100	[45]
Agave americana	218	652	2.99	1360	[46]

From the TGA results, the main degradation process of the hemicellulose and cellulose materials was observed to develop at approximately 230 °C, after some limited loss of non-structural matter and moisture. As shown in Figure 3, degradation mainly occurred between 324 °C and 380 °C, after which only some limited and gradual further decomposition took place. It is noteworthy that the degradation pattern of PCLF quite closely matched that of untreated pineapple leaf fibers (PALF) with regard to the temperature profile, as reported in [47], although the latter resulted in a residue of approximately 20% at 700 °C, higher than in the present case. In general terms, the stability of PCLF can be considered sufficient for use in traditional thermoplastics such as polyolefins, where processing can involve temperatures of over 200 °C.

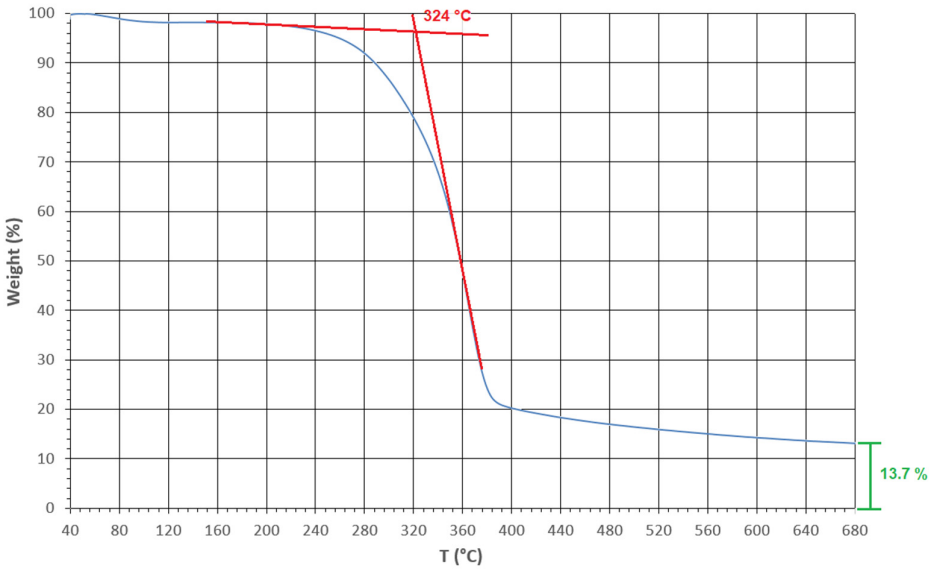

Figure 3. Thermogravimetric analysis (TGA) of PCLF.

A comprehensive FTIR spectroscopy study on pineapple leaf fibers (PALF) is offered in [48], from which some indication of the similarities and differences with PCLF can be elicited, as from the spectrum in Figure 4. While there are common absorption bands related to the activity of hydroxyls between 3480 and 3150 cm^{-1}, and C-H stretching between 2970 and 2800 cm^{-1}, some differences between the PALF and PCLF spectra

are present. The other peaks can be referenced as follows [48]: 1728 cm^{-1}, C=O stretching of hemicellulose; 1639 cm^{-1},possibly absorbed water; 1434 cm^{-1},O-H in plane bending of cellulose; 1322 cm^{-1},O-H bending and C-O stretching, which is unusually prolonged down to the peak at 1244 cm^{-1}—this is also attributed with respect to data indicated in [49]. The large peak at 1029 cm^{-1} was associated with C-O/C=C stretching vibration, as confirmed e.g., in [50], and that at 556 $cm^{-1}$with the torsional vibration of pyranose ring.

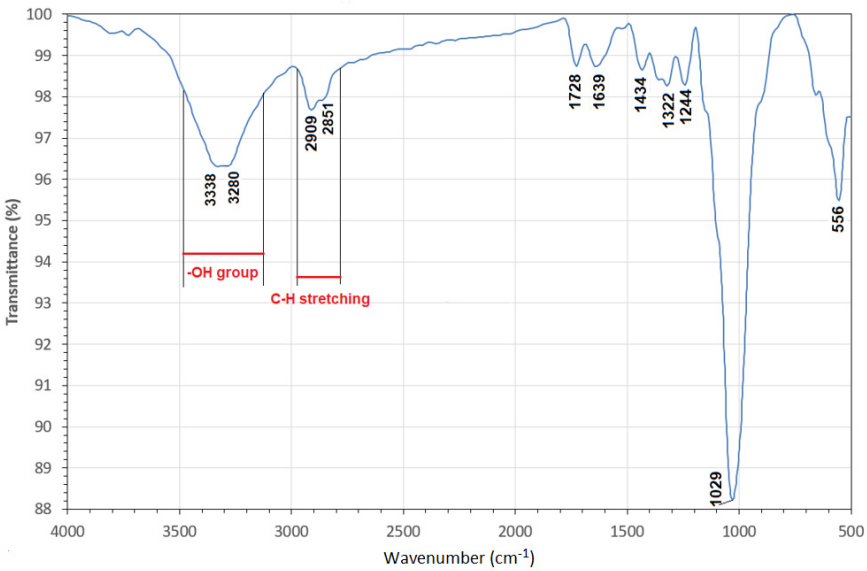

Figure 4. FTIR spectrum for PCLF.

The X-ray diffraction results allowed the calculation of the crystallinity index (CI) of the fibers, using the peak height method, as suggested in [51]. The peaks obtained, after baseline correction, corresponded to the different forms of crystalline native cellulose I, as is also normally the case in PALF, as thoroughly investigated for 12 cultivars in [52]. The measured peaks (Figure 5) indicated four crystalline reflections with relevant maxima, namely (101) at 14.8°, (021) at 19.7°, (002) at 22.4°, and (040) at 35.8°. The crystallinity index was calculated according to Equation (1), based on the intensity of the (002) peak I_{002} and of the low intensity peak I_{am}, corresponding to the amorphous cellulose fraction and localized at 17.7°, as first proposed in [53]:

$$CI = [(I_{002} - I_{AM})/I_{002}] \times 100 \quad (1)$$

The values obtained were compared with those measured for various other fibers, as from data reported in Table 3. It is notable that the properties of PCLF were very similar to that of untreated PALF, as reported in [54,55], while the crystallinity was lower in [56], which was suggested to be caused by the limited degumming achieved after mechanical retting. Larger differences are obviously observed compared to the other leaf fibers, although PCLF appears to be among those showing the highest crystalline indices.

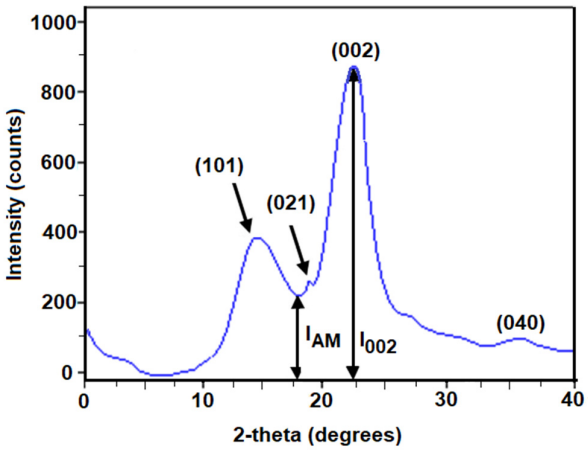

Figure 5. X-ray diffraction (XRD) spectrum for PCLF.

Table 3. Cellulose peaks and crystalline index as from XRD for different fibers (including PCLF)

Fiber	Peak Cellulose (101) (deg)	Peak Cellulose (002) (deg)	Crystallinity Index (%)	Ref.
PCLF	14.8	22.4	75.9	This study
PALF	15.1	22	76	[54]
Subang PALF	15.6	22.5	75	[55]
Queen PALF	15.5	22.9	52.2	[56]
Saharan aloe vera	-	22.6	52.6	[57]
Buriti (*Mauritia flexuosa*)	-	21.7	63.1	[58]
Furcrea foetida	15	22.6	52.6	[59]

Scanning electron microscopy (SEM) observations produced evidence that the untreated PCLF are not particularly prone to fibrillation when unloaded, as shown in Figure 6a. Further study at a higher magnification also provided evidence that, especially in the midrib areas, the fibrils can be folded at angles and twisted with different orientations (Figure 6b). It is noteworthy that pineapple leaves develop a midrib only at the later stages of their growth [60]. It is therefore likely that this fibril folding phenomenon would arise only once the crown is removed from the fully ripe fruit. On the other hand, retting did not allow complete degumming, as suggested by Figure 6c and d; the latter image also offers some indication of the rather repeatable and uniform diameter along the length of the fibrils. This is not the case for PALF, where variation in the diameter of the retted fibers up to a factor of 3 was measured on a sample of 120 fibers [61]. A more thorough investigation into the variability in diameter for PCLF will be the subject of further study.

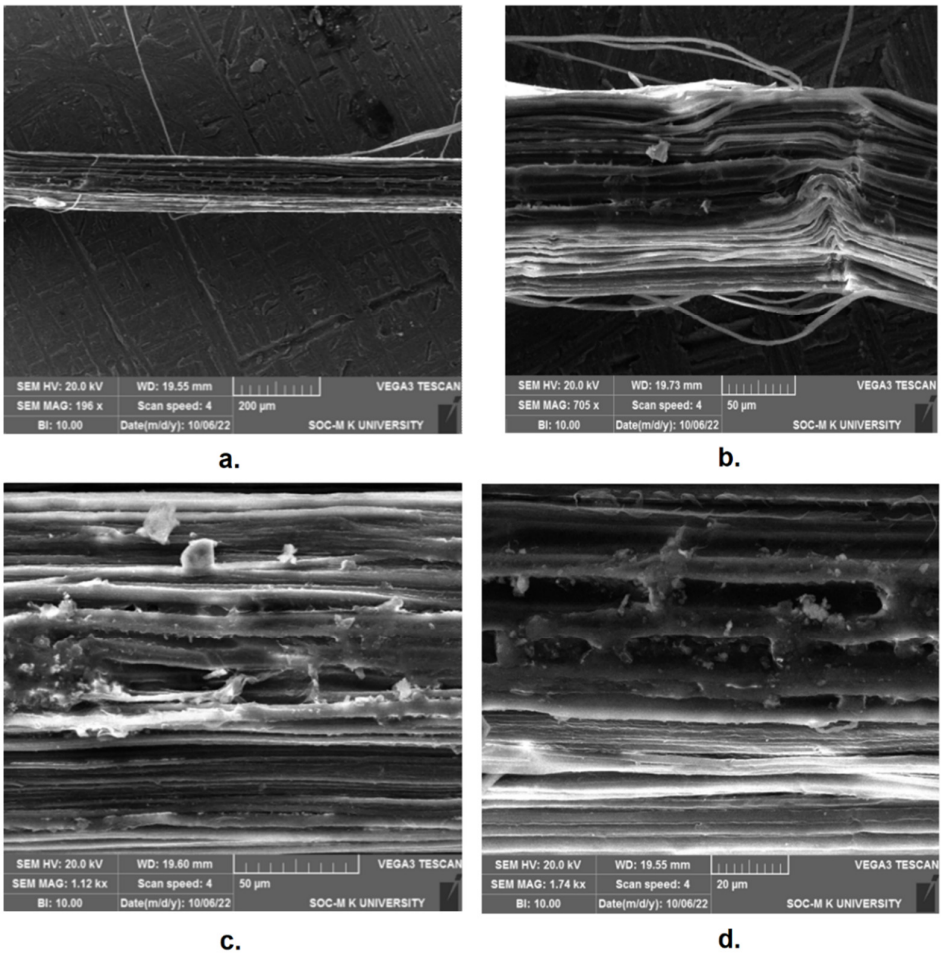

Figure 6. SEM micrographs of PCLF at different magnifications: (**a**) 196x; (**b**) 705x; (**c**) 1.12 kx; (**d**) 1.74 kx.

Atomic force microscopy (AFM) allowed the measurement of the roughness using a micrograph taken from PCLF; the profile is depicted in Figure 7, and the relevant data is reported in Table 4. It should be observed that the roughness measured was much larger than that found in previous studies on PALF (Malaysian Yankee cultivar), which might be attributed to the comparably limited dimensions reached by PCLF. In practice, values equal to 0.00356 µm, −0.0756, and 2.78 have been obtained for R_a, skewness, and kurtosis, respectively [62]. Another factor of concern might be the somewhat elevated ratio between R_q and R_a in PCLF, which was approximately 1.40, whereas close values of R_q and R_a would indicate a limited importance of steep and abrupt peaks in the roughness profile, hence a better approximation to a Gaussian distribution of peaks. This does not appear to be the case here compared with other untreated fibers, such as areca [63], where this ratio is equal to 1.19. Skewness, an asymmetry factor, was positive and higher in PCLF than in the other leaf fibers; an example of this can be seen in a study on *Dracaena Reflexa*, where a negative value of 0.875 was encountered, which was considered to be an indicator of high fiber porosity [64]. Kurtosis, the sharpness of the main peak, displayed a very high value compared even to fibers considered to be coarse, such as that shown in a recent study on *Echinochloa frumentacea,* where R_{ku} was measured as equal to 6.115 [65]. In general, the indications that were elicited from the roughness measurement for PCLF suggested a fiber with low porosity and highly delineated surface peaks. It is suggested that this could be improved by chemical treatment. On the other hand, it could be suggested that the higher roughness of PCLF might represent a positive factor for the fabri-

cation of composites, as it could offer stronger anchorage points for the resin, especially when, as would normally be the case for PCLF, short fibers would be used [66].

Table 4. Roughness parameters definitions and values.

Parameter	Definition	Value
Rp	Maximum peak height of the roughness profile	6.996 µm
Rv	Maximum valley depth of the roughness profile	3.183 µm
Rz	Maximum height of the roughness profile	10.179 µm
Rc	Mean height of the maximum profile elements	7.122 µm
Rt	Total height of the roughness profile	19.129 µm
Ra	Arithmetic mean deviation of the roughness profile	1.344 µm
Rq	Root mean square (RMS) deviation of the roughness profile	1.877 µm
Rsk	Skewness of the roughness profile	2.676
Rku	Kurtosis of the roughness profile	20.468
Rmr	Relative material ratio of the roughness profile	0.392%
Rdc	Roughness profile section height difference	1.989 µm

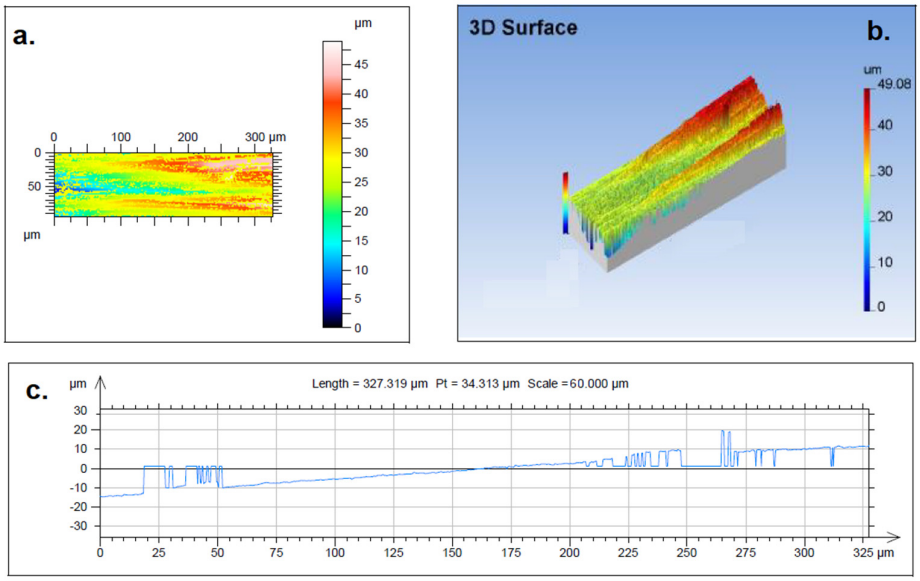

Figure 7. Roughness profile of the surface of one of the PCLF: (**a**) Upper view of the fiber; (**b**) 3-D view of the fiber; (**c**) Profile along one section line of the fiber.

Finally, tensile testing on the fibers offered a maximum load of 0.28 (±0.15) N, which, when considering the diameter as constant along the fiber, offered a maximum strength equal to 24.3 (±8.4) MPa, with a corresponding ultimate strain equal to 2.19 (±1.06) %. It proved difficult to apply the load, as illustrated in Figure 8, with the frequent load drops likely to have been due to torquing of the fibrils during loading, leading to their mutual detachment. This can possibly be attributed to the complex alignment of the fiber in the direction of the clamps which was due to a considerable fineness (in the order of 10 microns) of the fibrils arising during loading, despite the fact that their cohesion appeared to be sufficient before applying the force. The values for the tensile stress were obtained by considering the fiber as circular with constant diameter and are equal to the values measured at five locations along it, which might have led to a degree of imprecision. Out of the 20 fibers tested, five of the curves are reported in Figure 9. These were considered to be the most reliable results, in view of the aforementioned difficulties in acquiring the

stress and strain information. In Table 5, the data measured for PCLF are compared with the tensile strength and ultimate strain of the other leaf-extracted fibers.

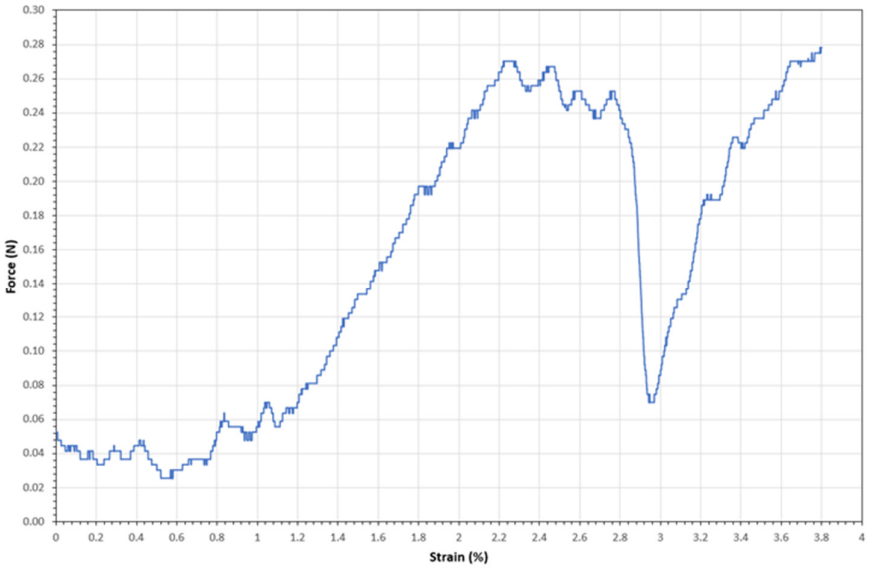

Figure 8. Typical tensile test curve on one of the PCLF.

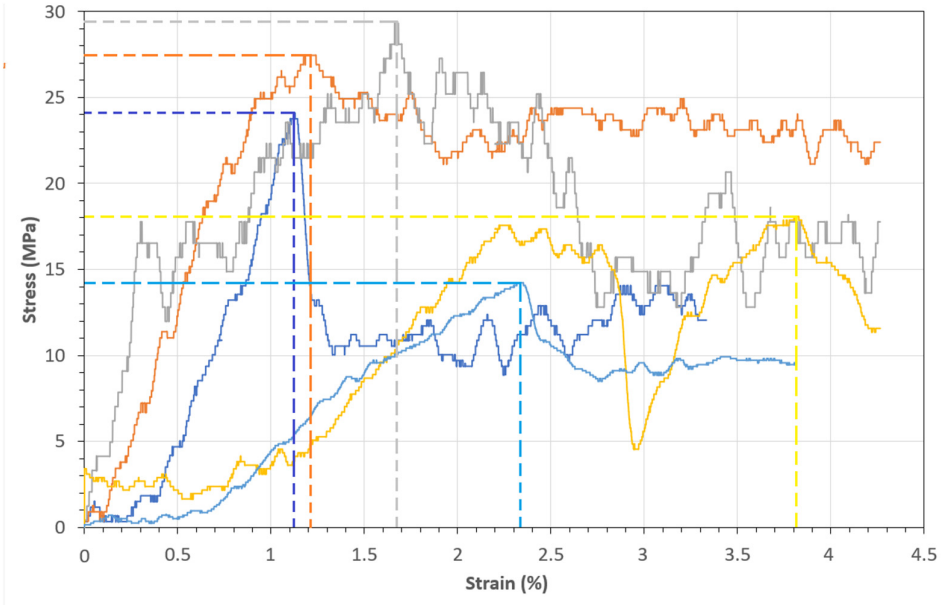

Figure 9. Some tensile curves for single PCLF.

Table 5. Tensile strength and ultimate strain of leaf-extracted fibers.

Fiber	Tensile Strength (MPa)	Strain at Max. Strength (%)	Ref.
PCLF	24.3 (±8.4)	2.19 (±1.06)	This work
PALF	630 (±50)	9 (±1.1)	[54]
Palm leaf	342	1.14	[67]
Echinochloa	204.32 ± 14.25	1.88 ± 0.11	[65]

In general terms, the characterization of pineapple crown leaf (PCL) fibers was aimed towards demonstrating the possible use of these as short fibers for composite re-

inforcement. This would allow PCLF to be separated from general pineapple waste. At the present time, if not immediately discarded, pineapple crown is disposed of in silage [68], or used for the extraction of bioactive compounds [69], bromelain in particular [70]. With a lower use profile, pineapple crown disposal by incineration and subsequent energy recovery is still a very common practice [71].

The scope of this work was to investigate whether using pineapple crown leaves (PCL) waste material as a short fiber source had merit. The use of pineapple leaf fibers (PALF) as short fibers e.g., for natural rubber reinforcement, has been widely investigated in the past [72,73]. Thus, the principal question that was posed was whether PCLF could compete with PALF, at least from a morphological, physical, and chemical standpoint. The investigation demonstrated that PCLF could compete with PALF, having approximately the same amount of cellulose, and therefore following a very similar degradation pattern, as observed in [74]. In addition, a degree of crystallinity was observed which was comparable to a number of other high crystallinity natural fibers, with a crystallinity index exceeding 70%, which was slightly higher than the data measured in [75] on pineapple crown from the Merryl cultivar in Brazil.

However, it is important to note that the effect of using different cultivars may be significant as far as variations are concerned; this has been observed comparing the results from various species of PALF [76], but has not yet been studied on PCLF, although the crown has considerably different geometries depending on the fruit cultivar [77]. The applications for PCLF also appear promising, owing to their limited tendency towards fibrillation and their relatively constant sectional dimensions compared to PALF. However, PCLF show higher roughness compared to most of the other leaf fibers studied, but this might provide a sounder interface with polymer matrices, especially in view of their application to stretches of a few centimeters.

4. Conclusions

This study on pineapple crown leaves fibers (PCLF), which specifically aimed to assess their application as short random fibers in composites, confirmed their substantial similarity to pineapple leaf (PALF) fibers, despite their limited dimensions. This similarity extended to their thermal and chemical properties, but less so to the morphological ones. Mechanical retting, although it did not result in effective degumming, offered fibers with a sufficient aspect ratio and limited proneness to fibrillation; furthermore, the PCLF were much rougher than PALF, therefore they are possibly adaptable to being used in different matrices.

Further development of this work would require investigating the possible need for treatment, which does not currently appear obvious, but needs to be assessed; this is also pertinent to the sustainability of the retting process, and to the optimization of the introduction of fibers into polymer or elastomeric matrices. The latter has been demonstrated to be particularly suitable for fibers of different lengths in PALF (normally in the order of a few centimeters, and to be adapted to the calendaring process), yet the advantage of using PCLF in this application would be to utilize waste product of the food production industry, rather than, as in the case of PALF, using a resource already used by other industrial sectors, such as the manufacturing of textiles.

Author Contributions: Conceptualization, S.P., M.P. and C.S.; methodology, S.P., A.K.M. and M.P.; investigation, V.J., V.K.R. and S.P.; resources, S.P.; data curation, V.J., V.K.R. and S.P.; writing—original draft preparation, S.P., M.P. and A.K.M.; writing—review and editing, C.S. and S.P.; supervision, C.S. and S.P. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Conflicts of Interest: The authors declare no conflict of interest.

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