Minimizing Organic Waste Generated by Pineapple Crown: A Simple Process to Obtain Cellulose for the Preparation of Recyclable Containers

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Abstract: In this study, cellulose was obtained from the residues of pineapple crown by means of simple acid pretreatment and subsequent alkaline treatment. The pretreatment consisted of washing, drying, and chopping with high shear at pH = 5 under heating. The content of cellulose, hemicellulose, and lignin in the pineapple crown was determined by chemical methods. The cellulose obtained was compared with commercial cellulose by Fourier-transform infrared (FTIR) spectroscopy, thermogravimetric analysis, and X-ray diffraction (XDR). Thus, from the obtained fiber cellulose, a food container was prepared, and its physical-mechanical properties were determined. Then, after alkali treatment, the purity of cellulose was 84.7% from the pineapple crown (56.0%) and was compared with commercial cellulose (95%). FTIR results confirmed the removal of the non-cellulosic compounds after alkali treatment. The maximum pyrolysis temperature increased to 356 °C, higher than the original fiber (322 °C), indicating greater thermal stability after chemical treatment. Furthermore, the crystallinity increased to 68% with respect to the original fiber (27%). The physical properties of the container showed a decrease in the parameters in wet 95% RH, as expected, thus facilitating its reuse. These results indicate that the pineapple crown cellulose can be obtained with significant purity, from a single chemical treatment. In addition, this polymorphous cellulose can be used to make ecofriendly reusable food containers.

Keywords: pineapple crown; cellulose fiber; high-shear homogenization; food container

1. Introduction

The pineapple (*Ananas comosus*) is the leading edible member of the family Bromeliaceae. This fruit is one of the most important in the world, and its juice is third-most preferred worldwide after orange and apple juices [1]. Around 10–25% of the total weight of a pineapple fruit corresponds to its upper crown [2]. Its processing generates 3 billion tons of by-products each year and has caused an environmental problem in agricultural lands [3]. Despite the technological development of the agricultural industry, plant and fruit parts are still discarded, wasting their potential economic uses [4].

Pineapple by-products generated by human consumption, commercial, industrial, or agricultural activity are a great source of contamination since these are not reused [5]. These by-products consist basically of residual pulp, peels, stem, and leaves. In general, these wastes have natural fibers, which are reusable as raw materials to obtain cellulose [6,7]. These natural sources are abundant and renewable,

and their reuse can solve adverse environmental problems [6]. Fibers, which can be obtained from the pineapple crown leaf, consist mainly of cellulose (79–83%), lignin (5–15%), hemicellulose (19%), pectin (1%), waxes (2–3%), and ash (1%) and have different physical properties [4,8–12].

There is an extensive body of literature that describes the use of fibers from the pineapple plant for different applications, such as in veterinary food [13], paper [14], clothing [15], cosmetics [16], and pharmaceutical industry excipients [16,17], among others [1,2,10,12,18–22]; however, few studies have used the fibers from pineapple crown waste [2,23–26]. This could be because the fibers of the pineapple crown are shorter than the fibers of the whole plant. However, despite pineapple plant fibers and pineapple crown fibers having similar composition and properties, many applications do not consider the fibers that can be extracted from pineapple crown.

Cellulose is the most abundant natural polymer on earth, and still is gaining attention due to its biodegradability, biocompatibility, renewability, availability, non-toxicity, chemical stability, and low-cost [2]. Cellulose is an unbranched homopolysaccharide with a partially crystalline structure, formed by a strong network of intramolecular and intermolecular hydrogen bonds, and is insoluble in water or other common solvents [27]. These important properties make cellulose very interesting to develop new biomaterials. In addition, this polymer has good thermal stability, resistance, rigidity, and low density [28,29].

Several sources of cellulose have been investigated, including prickly pear peel, banana peel, pomegranate peel, and palm trunk, among others [7,30,31]. There is an extensive amount of literature for obtaining cellulose from these sources through well-described methods, the most common being alkali treatment, bleaching, and acid hydrolysis [31,32]. Alkaline treatment disrupts the inter- and intramolecular hydrogen bonds between the hydroxyl groups of hemicellulose and cellulose, increasing the hydrophilic character of the polymers [30,32]. Acid hydrolysis with mineral acids can increase the crystallinity and size of the fibers [2,33].

Plastic is an easy-to-mold material, strong and easy to produce, but its residue is a serious global problem today [34]. In contrast, cellulose is a biodegradable and non-toxic alternative that could be an ecofriendly substitute for the manufacture of utensils that are mostly made of plastic [2]. Besides, cellulose can be used as reinforcement in polymeric matrices with hydrophilic groups, such as starch, polyethylene oxide, or polyvinyl alcohol. In these cases, utensils may be produced by thermoforming, and lower processing temperatures are required compared with the manufacturing of polyethylene-based utensils. Some of these matrixes such as starch are biodegradable, which allows the production of materials with several environmental benefits that can be used in applications such as in packaging, for example [1,2].

In Peru, only 1.9% of total reusable solid waste is recycled, such as paper, cardboard, plastic, metal, or glass [35], organic solid wastes simply being discarded. In Mariscal Nieto (Moquegua Region), an emerging city, fruit residues, particularly pineapple, are frequently discarded, which squanders a great source of natural fibers that could be reused after simple and inexpensive methods. In this work, the aim was to extract cellulose with a single chemical treatment in acidic conditions of the residues of chopped pineapple crown, thus obtaining a fiber with minimal amounts of lignin and hemicellulose. Furthermore, with the cellulose thus obtained, an ecofriendly food container was prepared. Cellulose was characterized by X-ray diffraction, thermogravimetry, and infrared spectroscopy. This simple process of obtaining cellulose from pineapple crown seeks to provide an alternative use of this natural fiber, which could also be commercially profitable for the inhabitants of the region.

2. Results and Discussion

The chemical composition of the fiber is displayed in Table 1. Pineapple crown *Ananas comosus* contained more cellulose, hemicellulose, and lignin before alkali treatment. After acid washing and alkali treatment, the percentages of lignin and hemicellulose were significantly ($p < 0.05$) reduced from 13.1% to 4.7% and from 16.8 to 6.8%, respectively. Furthermore, a significant ($p < 0.05$) increase in the amount of cellulose from 56.0% to 84.7% was observed. These values were in agreement with previous

data for the composition of *Ananas comosus* crown [1,2,36]. However, these values will depend on the variety and environmental factors where the fruit is grown [3].

Table 1. Chemical composition of pineapple crown before and after alkali treatment.

Treatment	Cellulose (%)	Lignin (%)	Hemicellulose (%)	Others (%)
PC	56.0 ± 5.2	13.1 ± 3.2	16.8 ± 2.2	14.1 ± 2.0
Alkali PC	84.7 ± 6.0 *	4.7 ± 1.0 *	6.8 ± 0.8 *	3.8 ± 0.4 *

* Values that are significantly different from the pineapple crown fiber (PC) (n = 4, one-way ANOVA, $p < 0.05$).

From the plant, natural fibers are extracted by mechanical and chemical procedures to remove different components in order to isolate the cellulose. The usual procedure, after cleaning raw fiber, consists of dewaxing with alkali treatment, delignification with a mineral acid, and subsequent bleaching [2,6,18,33,36]. In this work, partial delignification (4.7%) and hemicellulose removal (6.8%) under strong stirring at 70 °C was carried out, taking advantage of the pineapple crown's own acid medium (pH 4.7–5.2). The alkali treatment was necessary to remove the extractive pectin and wax. Acid hydrolysis with mineral acids was not necessary because this procedure can reduce the size of cellulose fibers and increase the crystallinity. Brittle materials would be thus obtained, which are less interesting for the preparation of food containers.

The physical appearance of pineapple fiber before and after alkali treatment is displayed in Figure 1. The brown color due to lignin, hemicellulose, and other substances has been reported by previous studies [2,24–26]. After acid washing and alkali treatment, the fiber becomes light brown (or cream). The removal of lignin and hemicellulose also can be visualized in the FTIR, XRD, TG, and DTG figures.

Figure 1. The physical appearance of crown pineapple fiber before (**A**) and after (**B**) chemical treatment.

Figure 2 displays the FTIR spectra of commercial cellulose, original, and treated pineapple crown fibers. The peaks around 3334 and 2910 cm^{-1} were assigned to O–H and C–H stretches of aliphatic moieties in polysaccharides [37]. The band at 1732 cm^{-1} was assigned to the ester linkage of the carboxylic group of ferulic and p-coumaric acids of lignin, suggesting considerable lignin removal after chemical treatment [37]. Characteristic peaks in the range of 1500–1600 cm^{-1} correspond to the aromatic skeletal vibration of lignin [38]. The typical C–H and O–H stretches are also presented in lignin, while vibration at 1604 cm^{-1} refers to the aromatic structure. Furthermore, the band at 1242 cm^{-1} is attributed to aryl C–O out of plane stretching vibration, which significantly decrease after chemical treatment [39]. The sharp peak appearing at 1638 cm^{-1} in all samples was attributed to the bending mode of water absorbed water in cellulose [37]. The peaks at 1163 and 896 cm^{-1} were associated with the C–O–C stretching β-(1→4)-glycosidic and C–H vibration of cellulose and hemicellulose [37,40]. In the same way, the peaks at 1022, 1313, and 1374 cm^{-1} are related to the C–O–C pyranose ring skeletal vibration, C–C and C–O skeletal vibration, and H-bonded OH group stretching, respectively, of cellulose [41,42].
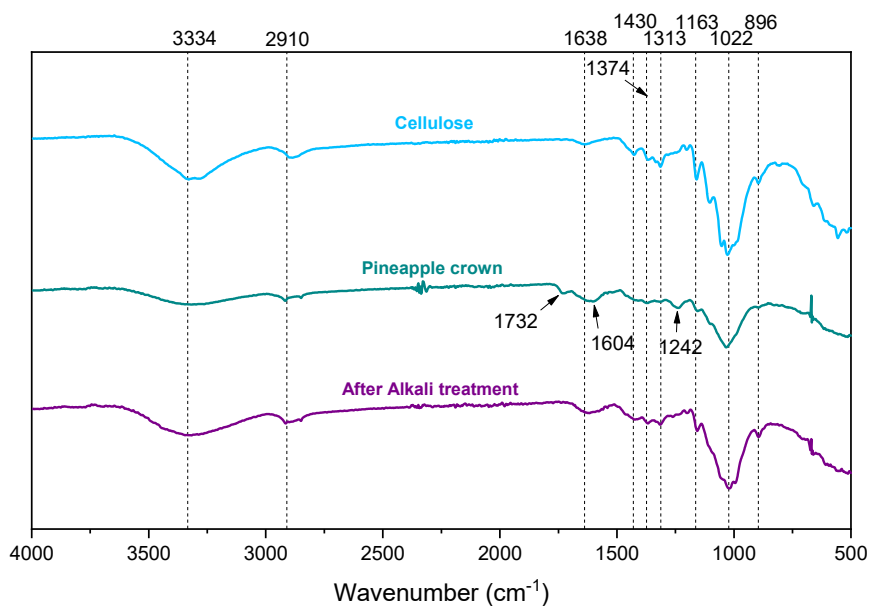
Figure 2. FTIR spectrum of the commercial cellulose, and before and after pineapple crown fiber treatment.

Cellulose, hemicellulose, and lignin usually decompose at different temperatures. The corresponding thermograms (TG and DTG) of commercial cellulose, original, and treated pineapple crown fibers are displayed in Figure 3. For all samples, a small weight loss occurs at below 100 °C, which can be attributed to absorbed water [43]. The commercial cellulose fiber decomposition started at 280 °C and persisted until 400 °C and the maximum weight loss rate was reached at 333 °C. At 400 °C, almost all cellulose was degraded, and the solid residuals were 4.3 wt.%. The pyrolysis of original pineapple crown fiber started at 136 °C and persisted until 450 °C, its weight loss rate (DTG curve) attaining maximum values at 164, 252.8, and 322 °C. The pyrolysis line from 136 to 300 °C indicates the decomposition of lignin-hemicellulose, while after 300 °C, the curve represents the decomposition of cellulose-lignin, mainly. Then, the decomposition curve after the chemical treatment of pineapple crown fiber started at 215 °C and persisted until 400 °C [43]. Furthermore, its weight loss increased greatly with the increasing temperature, reaching its maximum at 356.8 °C and split pseudo-Voight line corresponding to the cellulose-lignin-hemicellulose [43].

XRD diffractograms of commercial cellulose and of pineapple crown fiber before and after chemical treatment are displayed in Figure 4. In the XRD pattern of commercial cellulose, the well-known diffraction peaks at 2θ around 14.5°, 16.5°, 22.5°, and 34.5° were attributed to the $(1\bar{1}0)$, (110), (200), and (040) planes, respectively, with a crystallinity of 85% [44,45]. For pineapple crown fiber, weak peaks (2θ around 15°, 20.6°, and 22.0°) were detected and its crystallinity was calculated (27%) [44,46]. For pineapple crown fiber after alkali treatment, the diffraction peaks at 2θ around 14.5°, 20.3°, 20.6°, and 22.0°, corresponding to the $(1\bar{1}0)$, (110), (020), and (200) planes, respectively, were attributed to polymorphous cellulose, corresponding to a crystallinity of 68% [46,47]. This indicates that a polymorph of cellulose was obtained from pineapple crown after own-acid hydrolysis and alkali treatment. The probable explanation was that during the hydrolysis process in its own acidic medium, the amorphous region was not efficiently attacked by the acid. Thus, the adequate rearrangement of the cellulose to obtain a high crystalline structure did not happen [47].
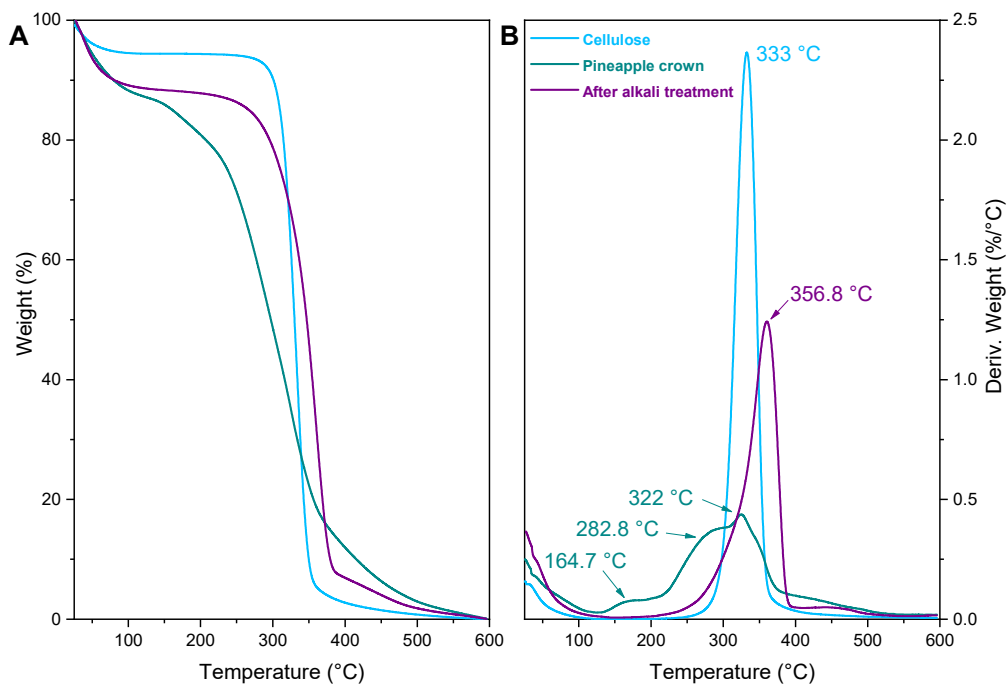
Figure 3. (**A**) Thermogravimetric analyses (TG) and (**B**) derivative thermogravimetric (DTG) analyses of the commercial cellulose and pineapple fiber before and after chemical treatment.
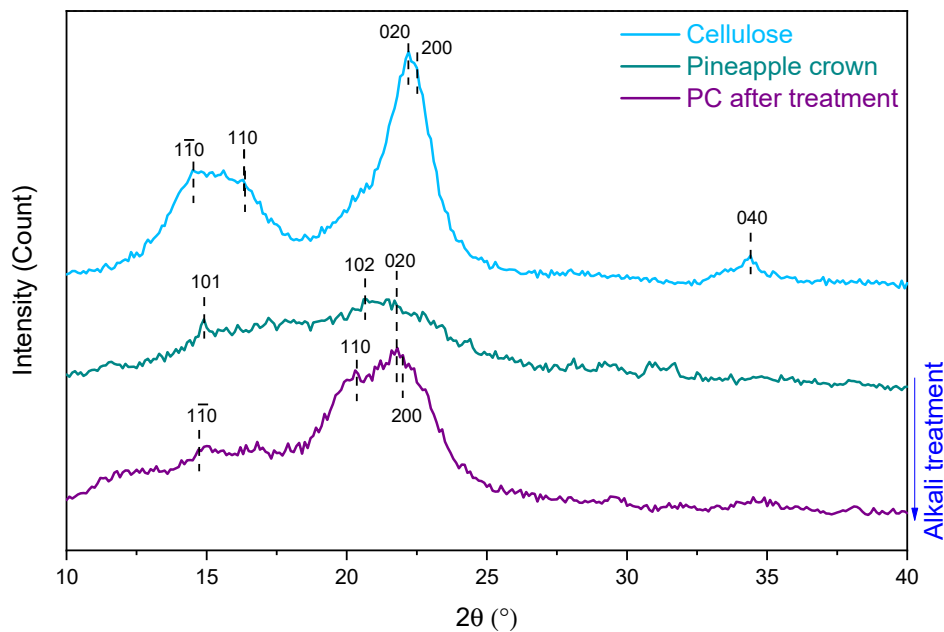

Figure 4. XRD patterns for commercial cellulose and pineapple crown fiber before and after chemical treatment.

Table 2 displays the physical-mechanical properties of the container (Figure 5), built as described in the "Mechanical proprieties" section. In general, as expected, increasing humidity significantly (* p < 0.05) reduced tensile stress (77%) and elongation (30%). The vapor interfered with the mechanical proprieties of the container, which can damage its elastic modulus and tensile stress drastically. Tensile strength is critical in a construction-related application. In contrast, the elongation (%) determines utility packaging. Our material has better properties than other commercial containers with similar utility, such as those made with Kraft paper (σ = 68 MPa, ε = 3%), corn starch (σ = 40 MPa, ε = 9%),

PEF (σ = 35 MPa, ε = 3%), or PS (σ = 30 MPa, ε = 1%) [6,48]. In addition, at room temperature (moisture = 20 ± 7%), our material displayed the necessary resistance for use as a food container.

Table 2. Physical mechanical proprieties of the container samples at dry and wet conditions (p = parallel, t = transversal).

Mechanical Proprieties	Dry Condition	Humidity Condition
Tensile stress (σ, MPa)	49 ± 2 [P]	11 ± 2 [P] *
	46 ± 3 [t]	9.2 ± 1.1 [t] *
Elongation (ε, %)	3.0 ± 0.2 [P]	2.1 ± 0.4 [P] *
	2.7 ± 0.3 [t]	1.8 ± 0.3 [t] *

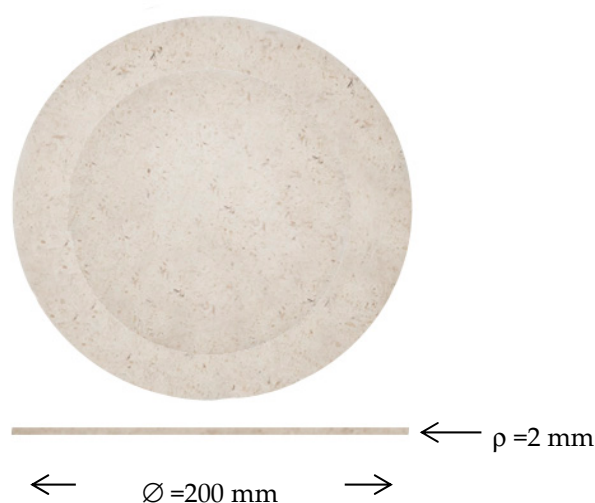* Values that are significantly different from dry condition (n = 4, one-way ANOVA, $p < 0.05$).

Figure 5. Container elaborated from pineapple crown cellulose (∅ = diameter, ρ = thickness).

The container's moisture was higher than the original pineapple crown fiber. This is due to the increase in the amount of cellulose and the addition of glycerin and starch in the formulation [36,49]. Indeed, the material has a large number of hydroxyl groups that strongly interact with water by hydrogen-bonding, increasing the affinity with water. Furthermore, the fragility or rigidity of the food container also will depend on several other inherent factors such as age, climate, and condition of the original plant that produced the fiber [50].

3. Materials and Methods

3.1. Chemicals

Cellulose (C6288, crystallinity = 85%, Sigma-Aldrich, St. Louis, MO, USA; 95%, medium fiber size), sodium hydroxide, pellets (Sigma Aldrich, ≥97%), starch from potato (Sigma-Aldrich, QL 100), and glycerol (Sigma-Aldrich, >97%) were used for the extraction of cellulose and container elaboration. All solutions were prepared using deionized water (18.2 MΩ cm^{-1} at 25 °C, total organic carbon ≤ 4.0 μg L^{-1}).

3.2. Cellulose Source

Pineapple crowns (*Ananas comosus*) were obtained in the free market from Mariscal Nieto, Moquegua, Perú, in 2019.

3.3. Cellulose from Pineapple Crown

Fresh pineapple crown leaves were washed with deionized water to remove dirt and dried in a laboratory oven at 50 °C for 24 h. Thereafter, the dried sample was mechanically pulverized and sieved to a particle size of 350 to 380 μm (final humidity 4–6%). A total of 40.0 g of pineapple crown powder was treated with 1.0 L of deionized water at 70 °C for 4 h, under strong mechanical stirring (pH 5.0–5.5). Then, the insoluble powder was collected and treated with 50 mL 10% (*w/v*) sodium hydroxide aqueous solution for 5 h at 70 °C under mechanical stirring. The alkali-treated fiber was washed with deionized water four times until the alkali was completely removed (washed thoroughly until neutrality) and then, it was dried at 65 °C for 12 h in an air circulating oven [51]. The yield of pineapple crown cellulose by the above processing was almost 28% (*w/w*).

3.4. Composition of Pineapple Crown Ananas comosus

The lignin content was determined according to adequate procedures described in the literature [52]. Pineapple crown (1 g, minced) was added to sulfuric acid (15 mL, 72% *w/v*) and maintained at 25 ± 1 °C for 2 h. Then, distilled water (560 mL) was added and the mixture was boiled for 4 h. The insoluble lignin was separated by centrifugation (10,000 rpm). The lignin was dried and weighed, and its quantity was determined using Equation (1):

$$Lignin = \frac{m_l}{m_i} \times 100 \tag{1}$$

where m_l was the obtained lignin mass and m_i was the initial sample mass.

Hemicellulose content was found by extracting cellulose from the holocellulose (hemicellulose + cellulose) according to procedures described in the literature [18]. Pineapple crown fiber (1 g) was mixed with acetic acid (1% *v/v*, 30 mL) and sodium chloride (0.3 g), and stirred for 1 h at 75 °C. The mixture was then cooled, and the residue was filtered and washed thoroughly with water. The residual solid was dried and weighed, and the quantity of holocellulose was determined using Equation (2):

$$Holocellulose = \frac{m_h}{m_i} \times 100 \tag{2}$$

where m_h was the obtained residue mass and m_i was the initial sample mass.

Cellulose content was determined by extracting holocellulose with sodium hydroxide (17.5% *w/v*) for 5 h at 25 ± 1 °C. The obtained powder was washed with distilled water (×5) until neutrality was reached. Then, the cellulose quantity was calculated using Equation (3):

$$Cellulose = \frac{m_c}{m_i} \times 100 \tag{3}$$

where m_c was the mass of white residue and m_i was the initial sample mass.

The tests were performed in quadruplicate.

3.5. Fourier-Transform Infrared Spectroscopy Analyses (FTIR)

FTIR spectra were recorded using a Fourier Bomem MB100 (Bruker, Billerica, MA, USA) spectrophotometer. Spectra were recorded from 4000 to 400 cm^{-1}, with a resolution of 4 cm^{-1}. The sample amount for each measurement was within 7–10 mg [51].

3.6. Thermogravimetric Analysis (TGA)

The thermal stability of the extracted fiber was investigated by a TGA Q500 V20.13 Build 39 (TA Instrument, New Castle, DE, USA) thermogravimetric analyzer. All the measurements were performed under nitrogen gas flux (60 mL min^{-1}). Each sample was kept in a platinum sample pan and

heated from 25 to 600 °C at a heating rate of 10 °C min^{-1}. The sample amount for each measurement was within 4–6 mg [45].

3.7. X-ray Diffraction Analysis (XRD)

The crystallinity of the fibers was investigated by X-ray diffraction (XRD) analysis using a powder X-ray diffractometer (Aeris XRD, Malvern Panalytical, Malvern, UK) with Cu Kα radiation (1.5406 Å) at 40 kV and 25 mA. The scanning range was from $2\theta = 5°$ to $40°$ at a scanning rate of $0.02°$ s^{-1}. The sample amount for each measurement was within the range of 300–500 mg. The crystallinity index (CI) was calculated from the intensity ratio of the crystallinity peak ($I_{200}–I_t$) and total intensity (I_{200}) [7] using Segal's method [53,54] (Equation (4)):

$$CI = \frac{(I_t - I_a)}{I_t} \times 100 \tag{4}$$

where I_t is the total intensity of the (200) peak for cellulose and I_a is the amorphous intensity at about (100)–(200) [53–55]. The average size of crystallite was calculated from the Scherrer equation (Equation (5)):

$$L = \frac{0.94\lambda}{H\, cos\theta} \tag{5}$$

where L is the size of crystallite perpendicular to the plane, λ is x-ray wavelength, H is the full width half maximum (FWHM, in radian), and θ is the corresponding Bragg angle [56].

3.8. Container Elaboration

To make one container, 20.0 g of cellulose from pineapple crown was added to 150 mL of distilled water and mixed under mechanical stirring (Blender Core, HR3573/91 Series 5000, Philips, Borehamwood, England, UK) until it reached the form of a homogeneous, slightly cream-colored paste (10 min, at 25 °C). Thereafter, 1.0 g of potato starch and 5.0 g of glycerin were added to the mixture, and the stirring was maintained for more 5 min. The final paste was placed, dried, and thermoformed at 200 °C for 10 min or in a tin cast at 150 °C for 30 min.

3.9. Mechanical Proprieties

Mechanical proprieties were determined as described [15,57]. A tensile testing machine (Instron 4520, Instron Corp., Norwood, MA, USA) was utilized to measure the mechanical proprieties of samples (initial length 25 mm, 4 mm, deformation rate 3 mm min^{-1}): tensile strength, elongation at break, and Young's modulus. The sample was measured in dry and wet conditions. For humidity conditioning, the sample was put in a chamber at 95% RH for 24 h. High humidity was obtained introducing boiling water inside the chamber along with the sample. Then, after humidification and measurements, the sample was promptly enveloped with a polyethylene film to prevent dehumidification. To determine the thickness of samples, a micrometer was used. The tests were performed in quadruplicate.

4. Conclusions

Cellulose has been successfully extracted from pineapple crown using a simple chemical treatment. The high-shear homogenization reduced the need for extensive chemical posttreatment. The main structure of cellulose was confirmed by FTIR, XRD, TG, and DTG tests, as compared with commercial cellulose. The crystallinity of the cellulose extracted from pineapple crown was increased, as was its degradation temperature (~360 °C). The cellulose extracted from the pineapple crown showed interesting properties that can be used in several applications with high added value. It was successfully applied in the preparation of ecofriendly food containers elaborated by mechanical thermoforming. Other potential applications for cellulose extracted from pineapple crowns can motivate the use of abundant and inexpensive by-products. Using the residues of the pineapple crown for the extraction

of cellulose appears as an important contribution to solve the problem of agro-industrial waste that pollutes our environment.

Conflicts of Interest: The authors declare no conflict of interest. The funders had no role in the design of the study; in the collection, analyses, or interpretation of data; in the writing of the manuscript, or in the decision to publish the results.

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