

Article

Comparative Study of Pretreatments on Coconut Fiber for Efficient Isolation of Lignocellulosic Fractions

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Abstract: Pretreatment is an essential step for breaking the recalcitrant structure of lignocellulosic biomass and allowing conversion to high-value-added chemicals. In this study, coconut fiber was subjected to three pretreatment methods to compare their impacts on the biomass's structural characteristics and their efficiency in fractionating the biomass. This comparative approach was conducted to identify mild biomass pretreatment conditions that efficiently extract lignin and recover cellulose-rich pulp for the production of bioproducts. To this end, autohydrolysis, alkaline, and organosolv pretreatments were performed under different experimental conditions, and the physicochemical properties of the samples were evaluated using scanning electron microscopy (SEM), Fourier transform infrared spectroscopy (FT-IR), thermogravimetric analysis (TGA), and chemical characterization of the cellulose, hemicellulose, and lignin fractions. Therefore, efficient experimental conditions were identified to pretreat coconut fibers with an extended understanding of the methods to process lignocellulose. Great delignification efficiency and pulp yield were obtained with organosolv > alkaline extraction > autohydrolysis under the selected conditions of 2 h at 185 °C in the presence of a catalyst, namely, 0.5 M NaOH, for 2 h at 55 °C and 20 min at 195 °C, respectively. FT-IR revealed a predominance of hydroxyl groups in fibers obtained from alkaline and organosolv pretreatment, showing higher lignin degradation and cellulose concentration in these samples. TGA revealed mass loss curves with similar behaviors but different patterns and intensities, and MVE analysis showed differences on the surfaces of each sample. The comparison of experimental parameters allowed the identification of suitable conditions for each extraction method, and structural analyses identified the specific characteristics of the fibers that could be obtained according to the method used. Therefore, the results are of great importance for developing sustainable and effective industrial processes.

Keywords: autohydrolysis; alkaline removal; organosolv; lignin; cellulosic pulp; coconut



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1. Introduction

The replacement of traditional feedstock with renewable resources for energy and chemical production coupled with the necessity of reducing CO₂ emissions from burning fossil fuels have shaped economic and social decisions for achieving sustainable development goals [1]. Within this context, the use of lignocellulosic biomass resources, which covers

a variety of plant-based biomass (crops, plant residues, forest resources, and dedicated energy plants) widely available at a relatively low price, has emerged as one of the most promising clean alternatives [2]. Lignocellulosic biomass is characterized by having a highly heterogeneity and complex chemical composition composed of cellulose, hemicellulose, and lignin, which are natural polymers and precursors of several bioproducts and chemicals of industrial interesting [3,4].

Cellulose is a biopolymer composed of monomeric D-glucose units linked in linear and unbranched chains by β -1,4 glycosidic linkages [5,6]. Hemicellulose is a branched polysaccharide polymer that contains a heterogeneous group of sugars monomers, mainly pentose units (xylose and arabinose) and hexose units (mannose, glucose, and galactose) and sugar acids, such as glucuronic and galacturonic acids [5,7]. Lignin, otherwise, is a high molecular weight and three-dimensional polymer with a phenolic chemical structure derived from phenyl propane units. Lignin is the second most abundant natural polymer on earth after cellulose [8,9].

Coconut, a fruit with an annual world production of 62.41 million tons, is an important source of lignocellulosic material [10]. The endocarp (inner shell part), mesocarp (fibrous part), and exocarp (outer shell) of coconut comprise about 60% of total fruit weight and are residues with high lignin and cellulose content that can be valorized as material for sustainable bioenergy and bioproducts [11,12]. However, coconut processing industries primarily focus on extracting coconut meat to produce oil, desiccated coconut, coconut milk, and other derivatives, as well as commercializing fresh coconut for water consumption. Unfortunately, they often neglect the residues, which remain unused, leading to multiple environmental problems [13–15].

In Indonesia, the Philippines, India, and Brazil, leading countries in the coconut market, contributing together to approximately 48.20 million tons of coconut production, the management of coconut solid waste has been a concern of environmental, social, and scientific relevance given the abundance that is currently generated and the prospect of expanding the market in the next years [10]. Therefore, the coconut industrial chain urgently needs to innovate the reuse and reintegration of its byproducts to maximize their value as alternative feedstocks for the industry and minimize the impact of pollution.

To obtain useful compounds for direct application or subsequent conversion into higher value-added chemicals and energy-dense fuels, coconut residues must undergo disintegration techniques that promote the breakdown of the lignocellulosic structural matrix and facilitate accessibility to their potential composition [16]. Pretreatments of a physical, chemical, and biological nature or their combination have been considered essential steps to favor cellulose, hemicellulose, and lignin depolymerization [17,18]. Pretreatment is performed to fractionate lignocellulose into its major components by removing one or more physical or chemical barriers that make raw biomass recalcitrant [19].

Recently, studies have investigated several approaches for the pretreatment of coconut residues, aiming to optimize lignin extraction and improve the accessibility and digestibility of cellulose. These methods include traditional techniques and alternative solvents, such as the use of deep eutectic solvents (DESs) and ionic liquids (ILs). Yierizam et al. [20] used DESs to evaluate the conversion of coconut husk for bioethanol, reaching a maximum lignin degradation of 16.97%. Using different ILs to treat coconut fibers, Gundupalli et al. [21] reached a delignification efficiency of 15.76 to 18.97%. Mankar, Pandey, and Pant [22] synthesized different DES and tested different experimental conditions for lignin extraction from coconut residue, finding the highest lignin yield of 82%. In another study, Anuchi, Campbell, and Hallett [23] fractionated coconut fibers using ILs, obtaining a lignin yield of 75% at 150 °C and 60% at 170 °C.

Common chemical treatments include dilute and concentrated acid hydrolysis, organosolv, alkaline, and autohydrolysis. Alkaline pretreatment is one of the most traditional means to disrupt cell walls, exposing cellulose and increasing surface area porosity. Depending on the severity, it may also cause the de-crystallization of cellulose [24,25]. The alkaline solution acts by dissolving acetyl and uronic acid in hemicellulose and degrading

ester and glycosidic side chains in lignin without degrading carbohydrates [26,27]. Schiavon and Andrade [28] reported a decrease in the lignin content of coconut fiber from 38.1% to 22.23–23.55% when using alkaline treatment. Autohydrolysis is an alternative chemical-free pretreatment that does not use any reagent besides biomass and any solvent or media besides hot water [29]. In this process, the effectiveness of the selective hydrolysis of hemicelluloses and partial extraction of lignin are influenced only by time and temperature parameters [24,30]. Carre et al. [31] obtained a lignin yield of 9.05% using an autohydrolysis–soda process.

Another alternative approach to lignocellulosic biomass treatment is the organosolv process, which uses organic solvents such as ethanol, methanol, propanol, glycerol, and acetone [32,33]. This process is widely employed and studied because of its high efficiency in delignification and removing hemicellulose while providing high recovery and accessibility of cellulose-rich pulp [34]. In the study by Avelino et al. [35], the authors obtained a lignin yield of 66% when applying the organosolv process.

Therefore, given the huge volume of coconut residues generated and the barriers still existing in accessing the structure of lignocellulosic biomass by traditional pretreatment methods, this study aims to evaluate different experimental conditions (time, temperature, concentration of reagents, and presence/absence of catalysts) of autohydrolysis, alkaline, and organosolv pretreatments toward the fractionation of the lignocellulosic structure in coconut fiber (CF). The objective is to compare different pretreatments and identify the most efficient condition of each in terms of lignin extraction and the yield and characteristics of cellulose-rich pulp for further conversion into value-added bioproducts.

2. Materials and Methods

2.1. Coconut Residue

In the current work, the green coconut mesocarp (coconut fiber) was selected as the lignocellulosic residue for investigation. Coconut fiber (CF) was collected from mature fruits acquired from an industrial unit located in Brazil's Northeast region. Prior to pretreatment experiments, fiber samples were washed, manually chopped to approximately 2 cm, dried, and homogenized.

2.2. Pretreatments

2.2.1. Soxhlet Extraction

Pre-processed CF was submitted to Soxhlet extraction to remove substances of lower molar mass. To do this, samples were placed in extraction thimbles and refluxed in the Soxhlet apparatus with 250 mL of ethanol P.A. for 4 h, with six cycles per hour. Then, cartridges and samples were dried in an oven at 50 °C for 24 h, and the obtained samples (named CFS) were homogenized and stored until further alkaline, autohydrolysis, and organosolv pretreatment, as shown in Figure 1.

2.2.2. Alkaline Extraction

For alkaline extraction, CFS samples were treated with variable concentrations of NaOH (0.25, 0.5, and 0.75 M) at a 1:25 solid-to-liquid ratio and for different reaction times (1 or 2 h). These experimental conditions were chosen based on previous studies [36,37], where the reported data were analyzed to define the optimal concentration and time range for efficient alkaline pretreatment under milder conditions. Each experimental run was performed at a mild temperature of 55 °C, and reactions were followed by cooling down and acidification with acetic acid to adjust the pH to 7. The recovery of treated fiber and liquors was accomplished using vacuum filtration. The solid pulp was washed with 150 mL of distilled water under vacuum filtration and oven-dried at 55 °C for 24 h. The filtrate was used for lignin determination.

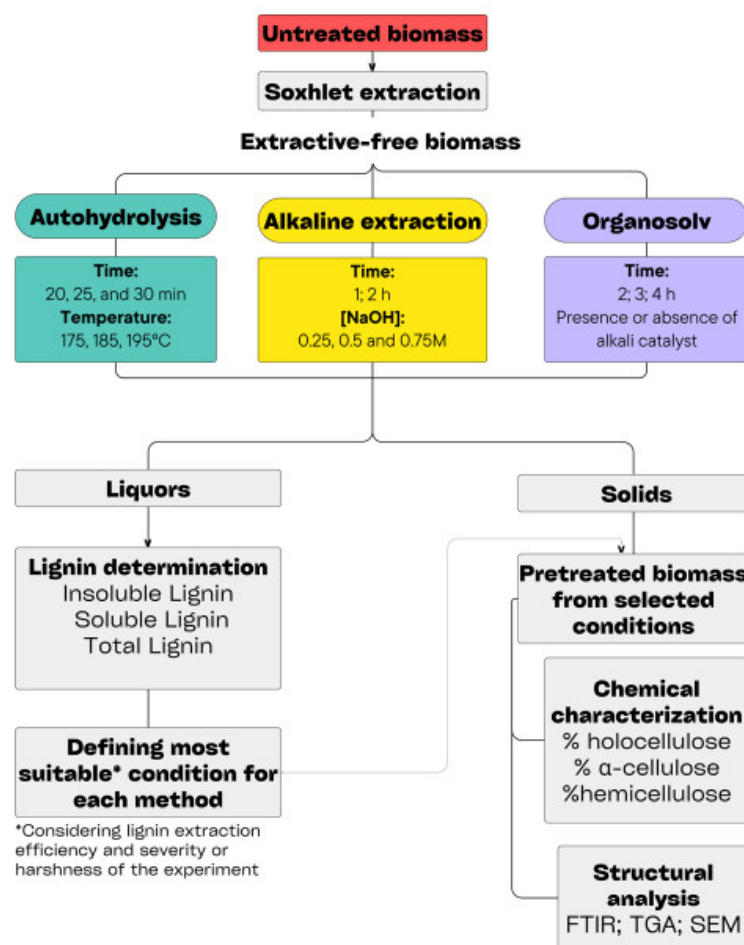


Figure 1. Flowchart of pretreatment processes applied to coconut fiber.

2.2.3. Autohydrolysis

The autohydrolysis process was performed in a stainless-steel reactor submerged in a silicone oil bath. In this process, CFS and distilled water were mixed at a 1:25 solid-to-liquid ratio, and experiments were conducted under variable conditions of time (20, 25, and 30 min) and temperature (175, 185, and 195 °C). Similar to the alkaline pretreatment, experimental conditions were discussed and chosen based on previous studies [30,38]. After the reactions, the reactor was cooled down in an ice bath, and the resulting mixture was separated using vacuum filtration. The solid residue was washed with 100 mL of distilled water, oven-dried at 55 °C for 24 h, and weighed to determine pulp recovery. The filtrate was used for lignin analysis.

2.2.4. Organosolv

For the organosolv treatment, the CFS was immersed in a 1:1 ethanol/water solution at a 1:25 solid-to-liquid ratio in the presence or absence of a 10% dry-weight alkaline catalyst (NaOH). The process was conducted in a stainless-steel reactor submerged in a silicone oil bath at 185 °C for 2, 3, and 4 h. After each run, the reactor was cooled down in an ice bath, and the pretreated biomass and liquor were separated using vacuum filtration. Solids were washed with 40 mL of ethanol/water, oven-dried at 55 °C for 24 h, and stored. The liquors were saved for lignin recovery and determination. Experimental parameters were adequately chosen based on the previous studies [31,39,40], establishing process conditions to evaluate the yield variation with time.

After the pretreatment experiments, all the liquors (filtrates) obtained were used for the analysis of insoluble (IL) and soluble (SL) lignin, and the results of total lignin (TL) extraction were used to determine the most suitable combinations of parameters for each

treatment. Then, fiber samples recovered under satisfactory conditions were characterized for chemical, structural, and morphological properties.

2.3. Biomass Characterization

2.3.1. Lignin Determination

Total lignin concentration was determined from the sum of SL and IL. IL content was obtained by the gravimetric method, following the procedure described by Kauldhar et al. [41] and Ismail et al. [42]. The solubilized lignin in each assay's filtrate fraction was precipitated by adding concentrated sulfuric acid until the liquor reached a pH of 2. The suspension was centrifuged, and the precipitated solids were washed with distilled water, followed by re-centrifugation to eliminate residual acid. The recovered solids were dried and weighed to determine the IL yield.

$$IL(g/L) = \frac{(1000M)}{V} \quad (1)$$

where:

M = mass of lignin precipitated in liquor (g).

V = volume of liquor aliquots used to precipitate lignin (mL).

The content of SL in the residual liquor was determined using UV-Vis absorption measurements on a UV-visible spectrophotometer from Bel Photonics, Model UV-M51, Valinhos/SP, Brazil. The absorbance was recorded at a wavelength of 280 nm, which was selected due to the affinity with the phenolic groups in the lignin structure [43,44].

$$SL_{280}(g/L) = \frac{A - 0.0313}{16.182} DF \quad (2)$$

where:

A = absorbance.

DF = dilution factor of liquor.

2.3.2. Chemical and Structural Characterization of Lignocellulosic Components

The pretreated fibers obtained from each experiment that were obtained under the most efficient conditions as determined based on the content of soluble and insoluble lignin were submitted for the characterization of holocellulose, cellulose, and hemicellulose. Fourier transform infrared (FT-IR) spectroscopy, thermogravimetric analysis (TGA), and scanning electron microscope (SEM) were used to evaluate structural properties.

Holocellulose content was determined according to the method described by Wise, Murphy, and D'addieco [45]. Samples were mixed with acetic acid and sodium chlorite and placed in a hot bath at 70 °C for four hours, with more acetic acid and sodium chlorite solution added every hour. For α -cellulose determination, 1 g of dried mass holocellulose was mixed with NaOH 17.5% *w/v*, added in two steps at different times and portions, and placed in a constant-temperature (20 °C) water bath [46]. Distilled water was later added, and the mixture was kept in the water bath for another 30 min. The mixture was filtered, washed with NaOH 8.3% and hot distilled water, and soaked in acid acetic followed by filtration. Once holocellulose and α -cellulose were quantified, hemicellulose content was determined based on the difference [47,48].

2.3.3. Fourier Transform Infrared Spectroscopy (FT-IR)

The analysis of the functional groups in the *in natura* (untreated coconut fiber) and pretreated CFS was performed using FT-IR spectrophotometer equipment equipped with a quartz surface (Model Spectrum Two, PerkinElmer, Waltham, MA, USA). FT-IR spectra of the samples were recorded in horizontal attenuated total reflectance (HATR) mode with a scanning range of 4000–450 cm^{-1} .

2.3.4. Thermogravimetric Analysis (TGA)

The thermal stability of *in natura* and treated CFS samples, previously dried in an oven at 50 °C for 24 h, was measured using a thermogravimetric analyzer (Thermogravimetric Analyzer, Model TGA-50, Shimadzu Corporation, Kyoto/Japan). For all analyses, the equipment was operated in a nitrogen atmosphere from room temperature up to 1000 °C at a heating ramp of 10 °C/min. A derivative (DTG) curve was used to portray the rate of the weight losses as a function of temperature.

2.3.5. Scanning Electron Microscopy (SEM)

The morphology of *in natura* biomass and changes caused by pretreatments were evaluated using scanning electron microscopy (SEM) (Hitachi, Model TM3000, Tokyo/Japan). Before analysis using SEM, samples were coated with silver and carbon to improve the conductivity and obtain approximate images.

3. Results and Discussion

3.1. Pretreatment Conditions

The concentration of extracted lignin in filtrates from pretreated assays was used as the main criterion to compare the different methods' performance and determine the best conditions among all tested for obtaining cellulose-rich pulp. High delignification capacity is related to the pretreatment's efficiency in disrupting the lignin–carbohydrate complex by splitting aryl ether linkages and solubilizing lignin, which increases cellulose accessibility. In this sense, maximizing lignin removal during the pretreatment step is essential to obtain cellulose fractions with a high yield and purity for the synthesis of different derivatives, such as methylcellulose, carboxymethylcellulose, and hydroxypropyl-methyl-cellulose [49]. Furthermore, the lignin recovered from pretreatment can also be exploited as bio-based material in several applications, such as biopolymers and biofuel production [50].

The CFS samples subjected to different pretreatments revealed a variable concentration of solubilized lignin in the liquors, in accordance with the employed method and processing conditions, as shown in Table 1.

Table 1. Lignin concentration in the pretreated coconut fiber liquors.

Autohydrolysis									
Temperature (°C)	175			185			195		
Time (min)	20	25	30	20	25	30	20	25	30
IL (g/L)	0.75	0.81	0.90	0.62	0.85	0.83	0.68	0.73	0.70
SL (g/L)	0.51	0.70	0.80	0.75	0.82	1.02	1.13	1.13	1.12
TL (g/L)	1.26	1.52	1.70	1.37	1.67	1.84	1.80	1.86	1.82
Alkaline extraction									
Time (h)	1			2					
[NaOH] (M)	0.25	0.50		0.75	0.25		0.50		0.75
IL (g/L)	1.41	2.99		3.54	1.84		5.89		6.72
SL (g/L)	0.93	1.22		1.53	2.82		3.81		3.69
TL (g/L)	2.35	4.20		5.06	4.66		9.71		10.41
Organosolv									
Time (h)	2			3			4		
Catalyst (NaOH)	P	A		P	A		P		A
IL (g/L)	93.10	6.30		126.10	7.40		155.56		43.60
SL (g/L)	6.84	2.52		5.91	3.75		5.42		2.09
TL (g/L)	99.94	8.82		132.01	11.15		160.98		45.69

IL: insoluble lignin; SL: soluble lignin; [NaOH]: concentration; P: presence of alkali catalyst; A: absence of the catalyst.

The total lignin recovered from pretreated liquors was calculated as the sum of the SL and IL fractions, in which SL refers to the small portion that is hydrolyzed into monomeric forms, and the insoluble portions is the condensed part, which generally accounts for most of the lignin in plant tissues. However, this study observed that the soluble portion of lignin was higher than the IL fractions for samples obtained from autohydrolysis at 185 °C for 20 and 30 min and at 195 °C for 20, 25, and 30 min and from alkaline extraction with 0.25 M NaOH for 2 h. This result highlights the importance of quantifying both fractions and indicates that the evaluated treatments may produce many soluble phenolic-derived oligomers with low amounts of condensed structures [51].

For the autohydrolysis pretreatment, it was observed that the lignin concentration increased with temperature and time, reaching maximum total lignin concentrations of 1.86 g/L at 195 °C/25 min and 1.84 g/L at 185 °C/30 min. Nonetheless, pretreatment at 195 °C for 20 min (1.80 g/L of TL) was selected as the main condition for further experiments since it produced results comparable to 25 min and 30 min. The increase in pretreatment time would imply extra energy consumption for a similar performance, making the selected condition, among all tested, the most convenient and viable.

For alkaline pretreatment, soluble, insoluble, and total lignin concentrations increased as the concentration of alkaline solvent and reaction time increased. As shown in Table 1, the total lignin concentration was greatly affected by the reaction time, which approximately doubled from 1 to 2 h, reaching maximum extractions of 10.41 g/L and 9.71 g/L at 2 h using 0.75 and 0.5 M NaOH, respectively. Once the difference in TL concentration between these two highlighted conditions was only 6.7%, the most suitable combination of parameters for the mild temperature of 55 °C involved the use of a NaOH concentration of 0.50 M for 2 h. Additional use of the alkaline reagent would not be beneficial in terms of treatment severity.

Regarding the organosolv pretreatment, the use of the catalyst significantly influenced the process, yielding lignin content at least three times higher than the corresponding non-catalyzed process (Table 1). The lignin concentration also increased with time, yielding maximum values at 3 and 4 h. Nonetheless, considering the effectiveness and treatment intensity criteria, the 2 h extraction condition was selected as a suitable pretreatment method because it showed high extraction efficiency but required less process time and energy consumption. Although alkali-catalyzed organosolv pretreatment for 3 h and 4 h showed superior extraction capacities, the lignin concentrations obtained under these conditions were not high enough to justify performing the process for an additional 1 or 2 h at 185 °C; thus, a moderate pretreatment intensity was preferable.

Based on the outcomes, it was perceived that an increase in temperature and time had a huge impact on improving the extraction of lignin. On the other hand, very long periods or excessive temperatures can stagnate or decrease extraction yield. In the study by Sujatha et al. [52], during the alkaline pretreatment of coconut fibers, the highest lignin extraction was observed at 120 min (320 mg/mL). Meanwhile, at 30 min and 150 min, lignin extraction produced yields of 275 mg/mL and 315 mg/mL, respectively. The authors attributed the decrease in extraction efficiency to lignin degradation when exposed to high temperatures and heat for long periods. Avelino et al. [35] otherwise did not observe significant alterations in lignin yield when varying the time from 20 to 30 min. However, an improvement in solvent performance was noted when the catalyst was added, which resulted in lignin yields of 56.6%, 54.4%, and 53.9% for H₂SO₄, HCl, and AlCl₃, respectively.

Ebrahimi et al. [53] determined the lignin concentration of coconut fibers *in natura* using the Klason method, reporting 2.51 g/L of total lignin, of which 2.212 g/L were insoluble lignin and 0.294 g/L were soluble lignin. For coconut fibers treated with an aqueous acidified glycerol-based organosolv process at 130 °C, total lignin yields of 2.42 g/L, 2.24 g/L, and 2.23 g/L were obtained with 15 min, 30 min, and 60 min of treatment, respectively. For the aqueous glycerol-based organosolv process under the same conditions, the concentrations were 2.42 g/L, 2.30 g/L and 2.21 g/L, respectively. It can be concluded

that low temperatures and shorter extraction times do not provide efficient delignification, as observed when comparing these results with the concentrations of lignin extracted using the organosolv process described in this study (Table 1).

Regarding the alkaline process, Gonçalves et al. [54] pretreated coconut fibers at 160 °C for 10 min with NaOH concentrations varying from 0.44 M to 1.6 M. The performed experiments highlighted the use of high temperatures and NaOH concentrations under shorter times, resulting in total lignin concentrations in liquors of 8.54 and 9.84 g/L, respectively. The tested conditions [54] provided results similar to those obtained in the present study (9.71 g/L) under the most efficient condition of 0.5 M NaOH at 55 °C for 2 h.

There is a gap in the literature on analyzing total lignin concentration in coconut fiber treated using autohydrolysis. However, Özyürek and van Heiningen [55] performed autohydrolysis experiments to pretreat wheat straw at 150 °C for 100 min. Outcomes revealed concentrations in g per 100 g of straw of 0.39 SL, 0.94 IL, and 1.33 TL. In the same study, the autohydrolysis pretreatment with varied concentrations of formic acid (2 g/L, 5 g/L, 10 g/L, and 15 g/L) provided total lignin concentrations of 1.66, 1.51, 1.53, and 1.60 g/100 g straw, respectively. These results indicate similarities in the progression of experimental conditions, leading to values of total lignin that were consistently close, as also observed in the results reported in Table 1.

3.2. Chemical Characterization

The treated samples recovered from each treatment carried out at selected processing conditions were characterized in terms of chemical content to compare treatment efficacy. As expected, all treated fiber showed lower lignin and hemicellulose content when compared to raw biomass, which increased from *in natura* > autohydrolysis $\approx$ alkaline > organosolv. It is worth mentioning that although alkaline pretreatment has provided a higher lignin extraction capacity than autohydrolysis (Table 1), the comparable percentage of retained lignin in a cellulosic solid is due to the calculation by difference considering the determined content of cellulose and lignin.

The organosolv process has been demonstrated to be superior in terms of total lignin removal and high cellulose recovery, which is attributed to the selective capacity to interact with lignocellulosic interlinkages, promoting greater fragmentation (enhanced by alkali-catalysis) and dissolution of coconut fiber lignin in the solvent [56]. On the other hand, the amount of lignin remaining in the pulp from alkaline and autohydrolysis suggested their lower selectivity to interact with lignin hydroxyl groups. Regarding the analysis of liquors using a US-Vis spectrophotometer, it could also be observed that organosolv has greater efficiency in obtaining soluble lignin, suggesting that the ethanol/water system protects lignin fragments from secondary condensation reactions. Highly condensed lignin is usually low purity, which greatly limits its application.

Regarding the concentration of holocellulose in coconut samples, higher yields were detected in pretreated fiber, which increased from organosolv > autohydrolysis > alkaline. The significant increase in carbohydrate content in fiber is mainly a result of the promoted depolymerization and solubilization of lignin, as evidenced by lignin concentrations in liquors and the removal of extractives. The higher concentration of alpha-cellulose (organosolv > alkaline > autohydrolysis) in the holocellulose fraction can be attributed to the removal of hemicellulose and other non-cellulosic components, leading to a more exposed alpha-cellulose structure.

Overall, it was noticed that all pretreatments showed similar capacity in removing hemicellulose, whereas the biggest difference between them was observed while removing lignin. The yield of recovery cellulose-rich pulp increases with the severity of the conditions employed in the pretreatment. The alkali-catalyzed organosolv process using 185 °C for 2 h resulted in fiber with 12% higher cellulose content than the obtained using alkaline extraction (non-catalyzed at 55 °C for 2 h) and 13% higher than autohydrolysis pulp (non-catalyzed at 185 °C for 20 min). Compared to the *in nature* sample, this increase was 28% greater.

The recovered fiber from alkaline extraction was found to have a cellulose yield 18% superior to non-treated CF and 3% higher than that obtained with autohydrolysis, which had 17% more cellulose than that noted for raw biomass. These findings show the importance of pretreatment conditions in obtaining materials with different chemical compositions, especially in terms of cellulose content. Analysis of lignocellulosic chemical characterization highlighted organosolv, among tested pretreatments, as the most effective to eliminate or cause a great reduction in the recalcitrance constructed by hemicelluloses and lignin in the one-pot process.

In Table 2, the levels of holocellulose, alpha-cellulose, hemicelluloses, and lignin found in the samples of this work are compared with results from other studies, reporting both *in natura* and treated fibers. The results exhibit different values but similar ranges and patterns. This divergence is attributed to several factors, such as variation in plants' genetics, cultivation, and climate conditions, and other environmental influences, as well as the methodologies used to determine the chemical composition (with or without the consideration of the presence of moisture and extractives) and the pretreatment conditions employed.

Table 2. Chemical characterization of raw and pretreated coconut fibers.

Residue <i>in natura</i>	Holocellulose (%)	α -Cellulose (%)	Hemicellulose (%)
This study	57.81 $\pm$ 0.5	40.01 $\pm$ 0.5	17.81 $\pm$ 0.5
Protásio et al. [57]	53.87	27.91	25.96
Liu et al. [58]	57.13	-	-
Parichanon et al. [59]	68.73	37.55	31.18
Alharbi et al. [60]	-	46.00	16.00
Gonçalves et al. [38]	-	32.18	27.81
Fleck et al. [61]	61.00	44.98	16.02
Autohydrolysis			
This study	58.37 $\pm$ 0.5	47.97 $\pm$ 0.5	10.40 $\pm$ 0.5
195 °C—20 min			
Gonçalves et al. [38]			
sequential NaClO ₂ -CH ₄ O ₂ /AH	77.92	71.25	6.67
200 °C—50 min			
Gonçalves et al. [30]			
200 °C—30 min	59.06	45.23	13.86
Alkaline extraction			
This study	58.08 $\pm$ 0.5	48.65 $\pm$ 0.5	9.43 $\pm$ 0.5
55 °C—2 h—0.50 M [NaOH]			
Schiavon and Andrade [28]	-	40.98	8.85
1 h—5% [NaOH]			
Schiavon and Andrade [28]	-	42.75	8.71
2 h—5% [NaOH]			
Bezerra et al. [62]	-	38.40	14.90
121 °C—30 min—0.50 M [NaOH]			
Din et al. [63]	-	40.14	12.64
121 °C—40 min—1.25 M [NaOH]			
Organosolv			
This study	65.91 $\pm$ 0.5	55.25 $\pm$ 0.5	10.66 $\pm$ 0.5
185 °C—2 h—ethanol:water:NaOH *			
Padilha et al. [64]	-	65.99	7.29
130 °C—2 h—glycerol:water:H ₂ SO ₄ *			
Nascimento et al. [65]	55.10	38.00	17.10
110 °C—20 min—acetic acid			
Nascimento et al. [66]	-	52.00	23.00
110 °C—3 h—acetic acid:HCl *			

*: catalyst.

3.3. FT-IR Analysis

As already discussed, the main structure of coconut fiber comprises cellulose, hemicelluloses, and lignin, which are predominantly composed of different functional organic groups such as aromatic, ketones, alcohols, and others [67]. The non-destructive technique of FT-IR spectroscopy was employed to characterize the chemical structure of the pretreated and raw samples. FT-IR absorbance peaks at specific wavelengths can reflect the changes in chemical content and profile caused by pretreatments, allowing the identification of the similarities and major differences between samples obtained using different extraction processes.

In this analysis, the functional groups were identified according to the literature data by associating each characteristic band of the spectrum with the corresponding functional group. The FT-IR spectra highlighting the most significant bands are displayed in Figure 2, and the most relevant groups in each pretreated sample are shown in Table 3. The spectra can be analyzed in two main regions: bands in the range of 3600–2800 cm^{-1} and bands that appeared at 1800–800 cm^{-1} (fingerprint region).

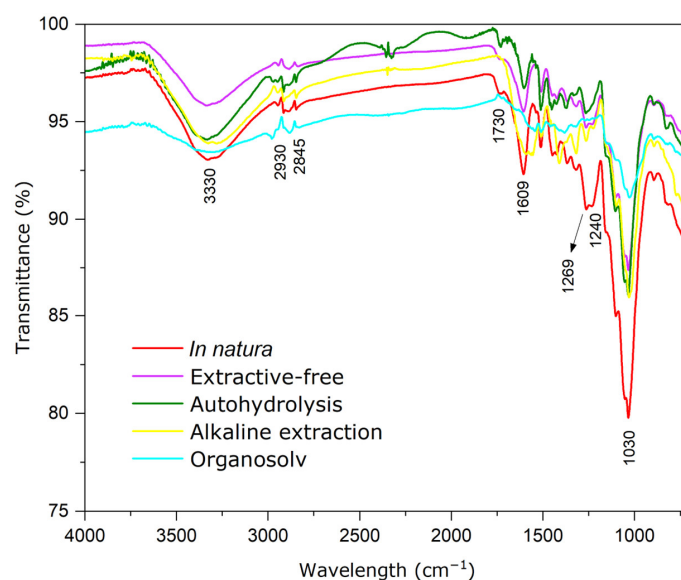


Figure 2. FTIR of *in natura* and pretreated coconut fibers.

Table 3. Bands assignments for coconut fiber FT-IR spectra.

Wavenumber (cm^{-1})			Functional Group	Chemical Structure
Autohydrolysis	Alkaline Extraction	Organosolv		
3337	3336	3336	stretching O-H	cellulose
2917–2851	2983–2920	2977–2884	stretching C-H	cellulose e hemicellulose
1730	*	1729	COOH; C=O	hemicellulose e lignin
1608	1604	*	linkage C=C	lignin
1512	1509	1509	aromatic ring vibration	lignin
1373	*	1380	deformation C-H	cellulose e hemicellulose
1328	1320	1320	vibration O-H	cellulose
1265	1266	1268	carbonyl groups C=O	lignin
1162	1162	1162	vibrations C-O-C	cellulose e hemicellulose
1035	1032	1028	stretching C-O	cellulose e hemicellulose
893	893	893	stretching C-O-C	cellulose

* did not show a peak near the specific band.

The broad peaks at 3600–3000 cm^{-1} correspond to the inter- and intramolecular stretching of OH (hydrogen bonds), which is due to the content of alcohol, phenols, and carboxylic acids that are present in all three macromolecules, namely, cellulose,

hemicellulose, and lignin [68–70]. The sharper and weaker bands in the range from 3000 to 2750 cm^{-1} are assigned to symmetric and asymmetric C–H stretching vibrations in CH, CH₂, and CH₃ groups, and are mainly attributed to the group's methylene and methyl that are also present in lignin and carbohydrates fractions [53,54]. The broad peaks in bands 3332–3330 cm^{-1} confirm the presence of cellulose in all samples. However, the lower intensity peak at 3332 cm^{-1} for treated samples indicate a slight disruption of O–H linkages in cellulose [71].

Vibrations around 2900 cm^{-1} are indicative of C–H in polysaccharides and the phenyl propane structure, which is observed for all samples. The peaks at 2938 and 2845 cm^{-1} are assigned to C–H stretching vibrations of hemicellulose and lignin, and the less sharp peak in the organosolv samples indicate a reduction in hemicellulose and lignin concentrations [72].

In the fingerprint region, bands at 1430, 1372, and 1336 cm^{-1} are characteristic of cellulose due to the bending of CH₂ (crystallinity band), C–H linkage, and bending in the plane of OH (amorphous), and the peak at 1317 cm^{-1} reflects the CH₂ wagging of crystalline cellulose [73]. The bands at 1430, 1373, 1337, 1319, 1161, 1107, 1058, and 1032 cm^{-1} are related to absorption of group C1, ring asymmetric stretching, C–O–C asymmetric stretching, and CH₂ symmetric bending vibration in native cellulose. Then, the low-intensity peaks reflect the change in the cellulose crystalline structure and an increase in the amorphous cellulose content [74,75]. At 1030 cm^{-1} , it could be observed that all samples had a remarkable peak, but with low intensity for organosolv pulp, which can be related to the change in the fiber structure caused by this treatment [73,76].

An analysis of cellulose purity in pulps can also be provided by comparing the specific bands wherein the absorption of lignocellulosic components does not overlap. The band at 1718 cm^{-1} is related to the presence of extractives, and a slight drop is noted in all treated samples. Bands at 1740–1730 cm^{-1} correlate to the carboxyl, ketone, carbonyl, and acetyl groups in hemicellulose, and a peak is noted for raw samples [71].

The bands near 1600 cm^{-1} are from the skeletal vibrations of the aromatic ring C=C present in lignin and COO[−] stretching from hemicellulose [68,77]. The peaks at 1595, 1530–1490, and 1463 cm^{-1} are mainly related to the skeletal vibrations of the aromatic ring C=C and C–H bending in CH₂ and CH₃ of the lignin structure [78]. From a broader view, it is possible to note a decrease in vibration intensity from organosolv > alkaline > autohydrolysis > extractive-free > *in natura*. Therefore, the lower intensity or absence of peaks indicates treatment efficiency in removing lignin and hemicellulose, which was observed mainly for organosolv samples [73]. This trend corroborates the chemical characterization results, wherein organosolv treatment promotes the reduction of hemicelluloses and lignin, and autohydrolysis and alkaline reduced hemicellulose content but has little effect on delignification.

The bands at 1264, 1240, and 1226 cm^{-1} are attributed to the C–O stretching in carboxylic acids and asymmetric stretching of arabinose chains in hemicellulose and the bonds C–O, C–H, and C=O due to the chain esters of the ferulic acid carboxylic acid group characteristics from lignin. For organosolv, the absence or smaller vibrations of peaks at these bands were observed, which is attributed to the absence or reduction of both lignin and hemicellulose [79]. Vibrations near 1200 cm^{-1} are attributed to the C–O–C or O–H-in-plane of carbohydrates; therefore, the lower intensity of treated samples compared to *in natura* CF is related to the removal of hemicellulose [74].

In summary, FT-IR analysis revealed that all samples showed peaks in the characteristic bands of each lignocellulosic component but presented different intensities for some of them. This finding confirms the presence of cellulose, hemicellulose, and lignin, showing that pretreatments led to more or less changes in the chemical content of CFS. Another important observation is that raw coconut fiber, Soxhlet-extracted coconut fiber, and alkaline-extracted coconut fiber samples have a slightly different pattern when compared to organosolv. According to the exposed, this difference can be explained by two factors: the high efficiency of organosolv in removing hemicellulose and lignin (as confirmed by the percentages displayed in Table 2) and the organosolv in the performed conditions showed some tendency to cause changes in

the cellulose structure. Nonetheless, this did not compromise the efficiency of pretreatment in increasing the content and accessibility of α -cellulose, which increased compared to the other treatments, as shown in Table 2. Also, a slight degradation of the cellulose structure can be beneficial for aiding in the hydrolysis of cellulose bioconversion [80].

Crystallinity Analysis of Samples Using FT-IR

When subjected to pretreatments, cellulose changes its hydrogen bonds, resulting in modifications to its physical and chemical properties and structural arrangements composed of amorphous and crystalline regions. For example, when treated with NaOH in the mercerization process, cellulose has an increase in its amorphous regions, and native cellulose (cellulose I) is partially converted into cellulose II.

The difference between cellulose types I and II arises from the organization and structure of molecules. Cellulose I, the native state found in plant fiber, has more crystalline regions and is characterized by cellulose chains ordered in parallel [3,81]. Cellulose II, a “swollen” polymorph of cellulose I, has an antiparallel arrangement and an increased presence of amorphous regions, which represents a non-crystalline phase of cellulose composed of non-ordered chains [3]. Due to these characteristics, type II has superior thermodynamic stability compared to cellulose I, making the conversion of cellulose I into cellulose II irreversible [82].

Characterizing the crystalline and amorphous regions allows us to visualize the changes and the influence of each pretreatment on the cellulosic structure of CFS. Once the characteristic absorption bands lignocellulosic bonds and components are identified using FT-IR, correlations between absorbance peaks at specific wavelengths can be used as a technique to identify and quantify the changes caused during the treatment of cellulosic materials because the spectrum of crystalline cellulose I and II and amorphous forms present different patterns [83,84]. In this sense, the empirical crystallinity indices total crystallinity index (TCI), lateral order index (LOI), and hydrogen-bond intensity (HBI) based on the absorbance at 1430 cm^{-1} , 1162 cm^{-1} , 1111 cm^{-1} , and 893 cm^{-1} were proposed to track the changes in cellulose crystallinity.

TCI is determined by the absorbance ratio at bands 1372 cm^{-1} and 2900 cm^{-1} ; the first is representative of crystallinity, and the second is not associated with crystallinity [84]. LOI represents the relationship between the intensity of the absorption band at 1430 cm^{-1} , characteristic of a crystalline region, and the band 893 cm^{-1} , typical of an amorphous region [85]. In contrast, HBI refers to the ratio between the bands 3400 cm^{-1} and 1320 cm^{-1} , which are related to the formation of hydrogen bonds between hydroxyls in cellulose, especially during the conversion of cellulose I to cellulose II, an increase in which implies a decrease in crystallinity [86,87]. In general, higher values of the TCI and LOI indexes suggest greater cellulose crystallinity and tend to be inversely proportional to the HBI [88,89]. Table 3 summarizes the band assignments for the FT-IR spectra.

Table 4 presents the crystallinity indices of the samples from this study. The samples subjected to Soxhlet and alkaline, autohydrolysis, and organosolv pretreatments showed LOI values higher than the raw CF sample, suggesting that the procedures reduced the number of amorphous regions. The changes observed can be attributed mainly to the removal of extractives and reduction in the hemicellulosic and lignin content (both amorphous polymers), which exposes the crystalline structures of cellulose. This behavior corroborates with the observed FT-IR analysis results presented in topic 3.3.1 of low or absence of peaks in bands characteristic of lignin and hemicellulose.

According to the work of Haykiri-Acma and Yaman [90], the effect of pretreatments on the fiber structure is directly related to its severity; thus, in milder experimental conditions, there is only the partial removal of lignin and hemicellulose, leading to a theoretical increase in the degree of crystallinity. However, when observing the reduction in TCI and the most significant increase in HBI for samples treated with organosolv, the results suggest that this technique not only caused the removal of lignin and hemicellulose but may also have led to the rupture of some hydrogen bonds, altering the cellulose structure, and making it more susceptible to hydrolysis. A similar behavior was observed for the values obtained

for autohydrolysis samples. However, due to the low concentration of lignin extracted compared to the other treatments (Table 1), the reduction in crystallinity is more likely to be related to the greater presence of lignin.

Table 4. Crystallinity indices of raw and pretreated coconut fiber samples.

Pretreatment	Index		
	HBI	TCI	LOI
<i>In natura</i>	0.8505	1.6206	0.8249
Soxhlet	0.9055	1.7606	0.8966
Alkaline	0.8303	1.6328	0.9372
Organosolv	1.2324	1.0076	0.8917
Autohydrolysis	0.4509	98.4246	0.7907

TCI: total crystallinity index; LOI: lateral order index; HBI: hydrogen-bond intensity.

3.4. TGA and DTA Analyses

Thermogravimetric (TGA) and differential thermal (DTA) analyses made it possible to evaluate the thermal behavior of the *in natura* and treated CFS. In all samples, mass loss with temperature occurred gradually and exhibited similar behaviors (Figure 3). The TGA profiles obtained in this study were similar to those reported by Fatmawati et al. [91] and Protásio et al. [57].

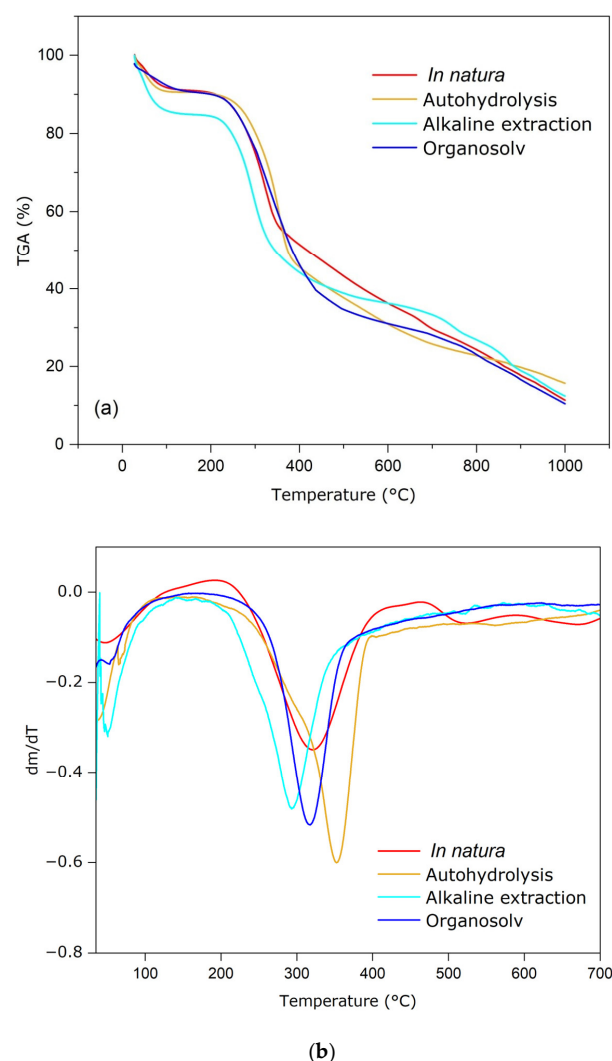


Figure 3. TGA (a) and DTA (b) of raw and treated coconut fiber.

The TGA and DTA curves portrayed in Figure 3 reveal that the thermal degradation of lignocellulosic biomass can be roughly separated into three regions (I up to 150 °C; II 150–450 °C; III >450 °C), and the process is perceived to occur as two major events. The first one even occurs in the temperature range of 27–120 °C, referring to the mass loss during drying, which is associated with the removal of surface water (moisture trapped in the biomass) and other volatile compounds. The second stage in the 200–400 °C range is linked to the effective degrading organic components, including hemicelluloses, cellulose, and partially lignin.

Lignocellulosic component degradation takes place at different temperature ranges and decomposition rates as the temperature increases. While carbohydrate backbones are broken more easily and over a narrow range temperature, lignin depolymerization occurs more slowly and continuously throughout the thermal process. This difference is related to the heterogeneity and complex chemical β -O-4 linkages in the lignin structure, which make the molecule more thermally stable and thus require more energy and time to break [92,93]. Consequently, residual solids are mainly attributed to lignin contribution [94]. Also, due to the branched and amorphous structure, hemicellulose is degraded at lower temperatures than cellulose.

Observing the mass losses in the temperature ranges of 27–120 °C and 200–400 °C, as shown in Table 5, it should be noted that the first recorded mass loss related to the presence of moisture and volatile components was similar for fresh samples (8.9%) and obtained from autohydrolysis (9.39%) and organosolv (9.10%) pretreatments. For alkaline extraction, the loss was 14.10%. The greater loss of mass in this sample compared to others can be attributed to three possibilities: (i) an indication that the alkaline process was not fully effective in removing volatile compounds; (ii) the presence of higher moisture content due to the increasing hydrophilicity caused by the break in OH bonds that create active hydroxyl groups in the fiber [95,96]; or (iii) the tendency of alkaline solutions to penetrate cellulose, leading to fiber swelling and increased amorphous regions. In the study by Singh et al. [97], the authors attribute the decomposition peak at a low temperature of 150 °C to the presence of a higher content of amorphous material in the fiber, which is less thermally stable and more easily broken.

Table 5. Mass losses from the TGA curves of coconut fiber samples before and after pretreatments.

Pre-Treatments	1st Mass Loss (%)	2nd Mass Loss (%)	3rd Mass Loss (%)
	27–120 °C	200–400 °C	>450 °C
<i>In natura</i>	8.90	38.90	36.02
Autohydrolysis	9.39	44.59	25.62
Alkaline	14.10	40.16	28.68
Organosolv	9.10	46.56	26.69

In the second stage, a more pronounced mass loss peak is obtained for all samples once the process is mainly associated with the thermal degradations of carbohydrates. The hemicellulose components react predominantly between 200–300 °C, and cellulose shows most reactivity between 250–350 °C, corresponding to the peak in the DTA curve. Lignin also has a partial contribution as it has the maximum peak of devolatilization in the range of 350–550 °C [94]. The mass loss in the temperature range of 200–400 °C was 38.9% for *in natura* samples and 40.16–46.56% for the treated ones, indicating effective degradation of hemicellulose and cellulose components.

In the TGA region above >450 °C, degradation is mainly related to the gradual lignin depolymerization and the carbonized materials [98], which explains the absence of a major event at any specific temperature. The greater mass loss of the sample subjected to alkaline extraction can be attributed to the characteristic of residual alkali lignin in cellulosic pulp. The lignin obtained by alkaline processes tends to be more condensed and contains high levels of ash, exhibiting resistance to thermal decomposition compared to other samples [99,100]. The coconut fiber sample submitted to autohydrolysis was found to have a higher content of ash than the other samples under the same temperature range of 1000 °C. This difference can be attributed to the preservation or formation of inorganic compounds

during the autohydrolysis process as well as the less efficient removal of impurities and organic matter, such as hemicelluloses and lignin. In addition, the decomposition of those components can lead to the formation of carbonized material, which contributes to the higher accumulation of solid residues [101].

These findings confirm the higher cellulose concentration in the treated sample due to the removal of hemicelluloses and lignin. As a consequence of lignocellulosic thermal behavior, samples with higher holocellulose and lower lignin content tend to have more mass loss in the second region, which corroborates the revealed outcomes.

3.5. SEM Analysis

Morphological analysis with scanning electron microscopy (SEM) was performed to track the foremost changes in the morphological structure of the fiber after pretreatments. Modifications were tracked by the presence of globular particles, the formation of cavities, and the absence of the layer that normally covers the fiber. In SEM images provided in Figure 4, it is possible to observe a rough surface for all fibers with the clearing presence of globular particles, suggesting substantial alterations in the surface layer influenced by the pretreatment processes.

The surface morphology of raw coconut fibers (Figure 4a,b) evidenced a layer comprising waxes, pectin, and other natural products that cover natural fiber. The resulting squamous texture with residual particles on the surface revealed the presence of pith or parenchyma cells, which are characterized by irregular shapes and clusters of dust, as observed in the provided SEM images [102,103].

Concerning fiber subjected to the Soxhlet process (Figure 4c,d), an initial exposure of globular particles could be noted while conserving the squamous appearance. The same pattern was noted in the fiber treated with autohydrolysis (Figure 4e,f), which exhibited a similar morphological structure. However, there was a greater presence of globular particles, indicating the removal of impurities deposited on the surface, extractives, and other compounds. These observations suggest that both processes drove a smooth change in the morphological structure of the coconut fiber.

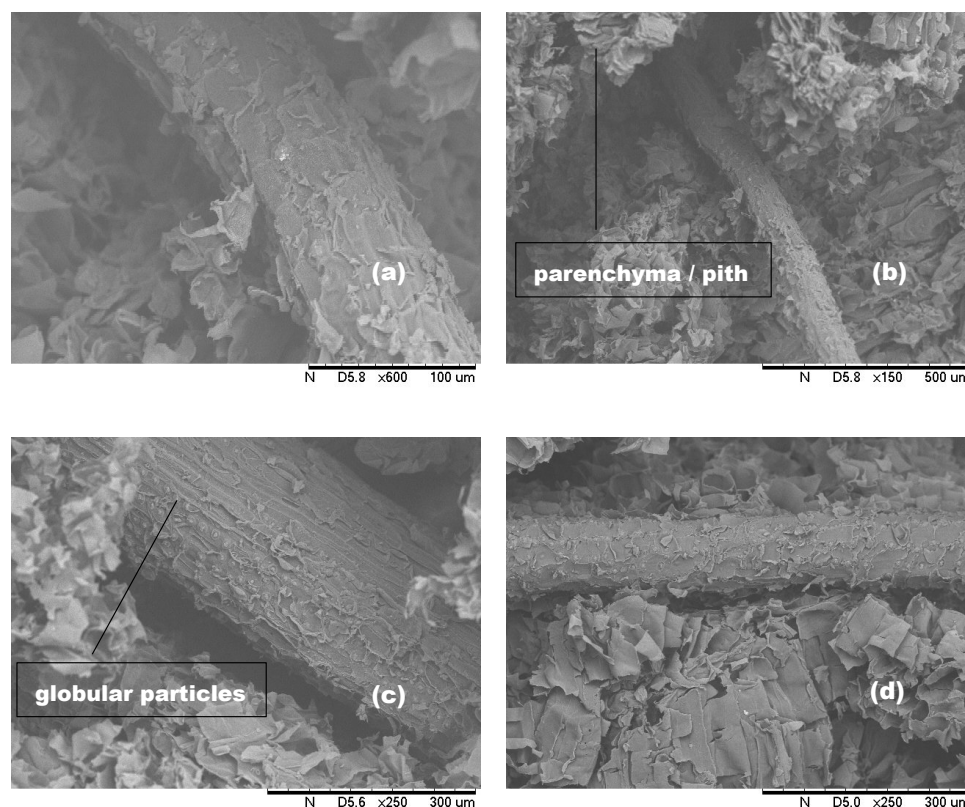


Figure 4. Cont.

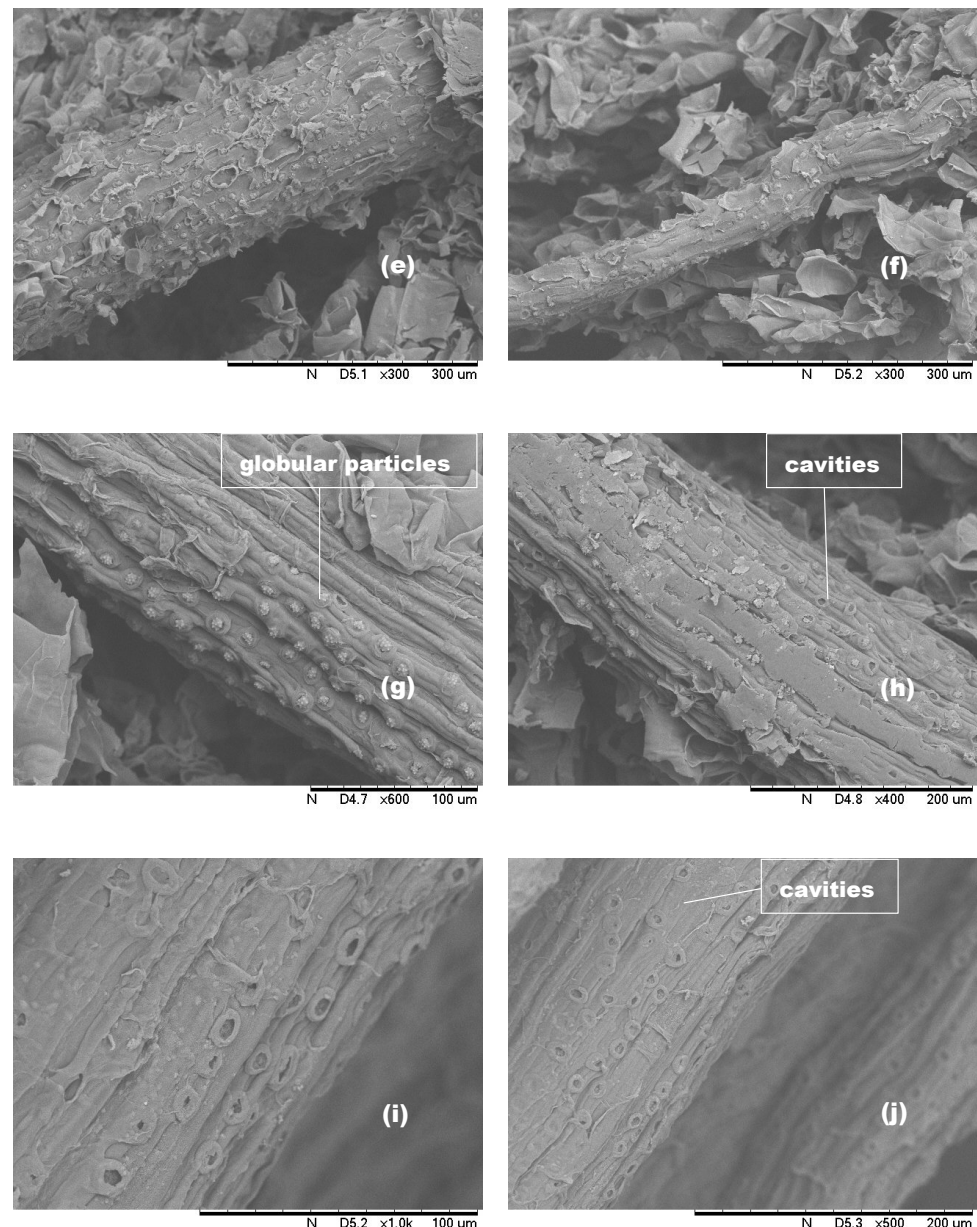


Figure 4. Scanning electron microscopy images of coconut fiber *in natura* (a,b); and chemically treated by Soxhlet (c,d); scanning electron microscopy images of coconut fiber: autohydrolysis (e,f); alkaline extraction (g,h); and organosolv (i,j).

SEM analysis helps confirm the effects highlighted in the previously discussed analysis. Alkaline pretreatment and the organosolv process aim to remove hemicellulose and lignin from biomass, exposing cellulose and some carbohydrates from residual hemicellulose. The removal of lignin and other constituents is related to the absence of the squamous layer associated with roughness and cavities, suggesting that these processes caused a more significant change in the fiber morphology. After the organosolv process, the sample exhibits a more intense roughness and the greatest number of cavities, allowing the conclusion that the organic solvent has a more effective action toward the breakdown of lignocellulosic matrix compared to alkaline extraction and the other tested pretreatments.

4. Conclusions

From a general analysis, the results demonstrate that all pretreatments tested could promote an efficient removal of hemicellulose and extractives, increasing the alpha proportion content in the holocellulose fraction. Nonetheless, their efficiency and the characteristics of the extracted components varied greatly among the processing conditions, specifically the performance of organosolv, which was deeply influenced by catalyst presence.

When evaluating methods' performances under their respective selected conditions of 185 °C and 2 h in the presence of catalyst (organosolv), 0.5 M NaOH for 2 h at 55 °C (alkaline extraction), and 20 min at 195 °C (autohydrolysis) and considering the criteria of lignin and hemicellulose extraction capacity and high yield of cellulosic fiber, the organosolv process was demonstrated to be the most effective pretreatment for coconut lignocellulosic biomass. Liquor from the alkali-catalyzed organosolv process was found to yield an outstanding concentration of total lignin, and the respective recovered pulp was found to have a higher cellulosic fraction, which suggested a lower content of residual lignin in organosolv-treated samples compared to alkaline extraction and autohydrolysis. The poorest efficiency regarding the lignin concentration in liquors was observed for autohydrolysis, which was expected since this method uses only water as a solvent. In terms of structural changes in the obtained fiber, organosolv was also highlighted as causing some slight changes in the cellulose structure, probably due to the severity of the process.

Findings from this study contribute to selecting pretreatment methods focusing on identifying a more efficient set of parameters for mild experimental conditions, aiming to improve the use of coconut fiber as feedstock for the bioconversion process. In addition, the chemical and structural characterization of each coconut fiber sample allowed us to determine the most efficient method and condition to isolate lignocellulose fractions. These aspects and the discussion highlight the novelty of this work. Although this work did not quantify the environmental cost impacts of the tested pretreatment system, it is suggested that a deeper analysis addressing energy and water consumption can aid in choosing the ideal method. Environmental and economic assessments of the three pretreatments can provide a broad view of their impact and effectiveness for future research and industrial applications.

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