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ORIGINAL ARTICLE

Extraction and characterization of cellulose microfibers from Retama raetam stems

Abdelkader Khenblouche[1]*, Djamel Bechki[1], Messaoud Gouamid[2], Khaled Charradi[3], Ladjel Segni[4], Mohamed Hadjadj[5] and Slimane Boughali[1]

[1]Laboratory of New and Renewable Energy in Arid Zones – LENREZA, University of Ouargla, Ouargla, Algeria

[2]Department of Chemistry, Faculty of Mathematics & Material Sciences, University of Ouargla, Ouargla, Algeria

[3]Laboratory of Nanomaterials and Systems for Renewable Energies – LaNSER, Research and Technology Center of Energy, Techno-park Borj-Cedria, Hammam-Lif, Tunis, Tunisia

[4]Laboratory of Process Engineering, Faculty of Applied Sciences, University of Ouargla, Ouargla, Algeria

[5]Laboratory of Valorization and Promotion of Saharian Resources – VPRS, University of Ouargla, Ouargla, Algeria

**khenblouche89@gmail.com*

Abstract

Cellulose is the most abundant renewable resource in nature, it has various industrial applications due to its promising properties. Retama raetam is a wild plant belonging to the Fabaceae family, largely abundant in arid area which makes it a good candidate for industrial utilization. In the present study, highly crystalline cellulose microfibers (77.8% CrI) were extracted from Retama Raetam stems as a novel renewable source. The samples underwent a dewaxing process, then the microfibers were extracted using 7 wt% sodium hydroxide followed by a bleaching treatment. The extracted cellulose microfibers were characterized by Scanning electron microscopy, Fourier transform infrared spectroscopy, X-ray Diffraction and thermo-gravimetric analysis.

Keywords: *cellulose, microfibers, Retama raetam, extraction, characterization.*

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1. Introduction

Over the last few decades, The use of natural fibers instead of synthetic fibers as reinforcement materials for polymer composites has gained considerable attention because of their unique characteristics, such as renewability, biodegradability, processing flexibility, low density, high specific strength and low-cost[1,2]. In addition, natural fibers have applications in various fields such as bioenergy industries, automobiles, paper manufacturing and textile owing to their properties and broad availability[1,3]. As a result, nowadays, the subject of many researchers worldwide focuses on the need to find alternative fiber sources[4]. Among all natural fibers, cellulose has attracted much interest as it is the most abundant renewable resource in nature and the degradation of cellulosic biomass is an important part of the biosphere's carbon cycle[5]. Its existence as the common material of plant cell walls was first investigated by Braconnot in 1819[6] and Payen in 1838[7]. It is a polydispersed linear polymer with a microfibrillar structure composed of poly-b (1→4)-D-glucose units with a syndiotactic configuration[8,9], found in the cell walls as a network of microfibrils embedded in a non-cellulosic matrix[10]. Several plants are rich in cellulose, i.e. cotton, wood, bamboo, hemp, flax, and jute …etc[11]. In addition, cellulose fibers have been extracted from several sources such as; milkweed stems[12], hop stems[13], rice husk[14], Cissus quadrangularis root[15], dichrostachys cinerea bark[1] and many more. Although several sources of natural fibers were investigated in detail, the isolation of cellulose fibers from R. Raetam has not been reported yet.

Retama raetam, locally known as R'tem, is a wild plant of the Fabaceae family. It is common to North and East Mediterranean regions[16]. It is largely abundant in arid area; this abundance makes it a good candidate for industrial utilization. Moreover, the Retama species contributes to the bio-fertilization of poor grounds because of their aptitude to associate with fixing nitrogen bacteria Rhizobia[17]. Therefore, the genus of the Retama is included in a re-vegetation program for degraded areas in semi-arid Mediterranean environments[18].

In this research, Natural micro-sized cellulose fibers were extracted from R. Raetam stems using alkali and bleaching treatments, the resultant cellulose microfibers were characterized using FTIR, SEM, XRD and TGA.

2. Materials and Methods

2.1 Materials

Stems of R. raetam subject of this study were collected in Ouargla, Algeria, in 2015. Acetone, Ethanol, Sodium hydroxide & Hydrogen peroxide were purchased from Sigma-Aldrich and were used without further purification.

2.2 Preparation of samples

Adult stems were cleaned with water and air dried, broken to the size of about 1 cm long and 1 mm width, grinded into powder with a Retsch SM100 Comfort cutting mill (Retsch GmbH, Haan, Germany), and sieved using a sieve size of 0.25 mm.

2.3 Microfibers extraction

The extraction of cellulose microfibers was performed using classical chemical treatments with adaptations in dewaxing, alkali and bleaching treatment processes. The totally chlorine-free extraction procedure can be described as follows:

2.3.1 Dewaxing

About 20g of powdered stems were first dewaxed in a Soxhlet reflux with a 2:1 (v/v) mixture of Acetone/Ethanol at 63 °C for 7 h, the main purpose of this step is to remove off waxes and extractives, the sample was then placed in a Buckner funnel and vacuum dried at room temperature for 3 h to remove traces of residual solvents.

2.3.2 Alkali treatment

The alkali treatment was performed to purify the cellulose by removing lignin and hemicellulose from R. Raetam fibers. The extractive-free sample was treated with an alkali solution (7 wt% NaOH) with a solvent to solid ratio of 10:1 at 80 °C for 3 h under mechanical stirring. This treatment was performed trice, after each treatment the solid was filtered and washed with distilled water until neutral pH.

2.3.3 Bleaching

A subsequent bleaching treatment was carried out to remove residual lignin and whiten the microfibers. The sample was immersed in a hydrogen peroxide solution (11%, v/v), the pH was adjusted to 11 using 7 wt% NaOH, the system was vigorously stirred for 3 h at 45 °C. For a more effective discoloration, the bleaching process was performed twice under the same conditions, after each treatment, the microfibers were filtered and washed with distilled water.

2.4 Fourier transform infrared spectroscopy

To analyze the chemical changes of the samples and investigate functional groups in the extracted cellulose we used Fourier Transmission Infra-Red Spectroscopy. The FTIR spectra were recorded on a Cary 660 FTIR Spectrometer (Agilent Technologies, USA) in a wavelength range of 4000–600 cm^{-1} with a resolution of 4 cm^{-1}.

2.5 X-ray Diffraction (XRD) analysis

The crystallinity of cellulose microfibers was investigated by X-ray diffraction (XRD) analysis, using a powder X-ray diffractometer (D8 Advance A25 Bruker AXS GbmH., Germany) with Cu Kα radiation (1.5406 A˚) at 40 kV and 25 mA, in the range of 2θ = 5-60° at a scanning rate of 0.02° s^{-1}. The crystallinity index (CrI) was calculated according to Segal equation[19]:

$$CrI = 100 \times \left(I_{200} - I_{am} \right) / I_{200} \quad (1)$$

where I_{200} is the diffraction intensity at 2θ = 22-23˚; and I_{AM} is the minimum diffraction intensity at 2θ = 18-20˚.

The crystallite size was calculated as per the Scherrer equation[20]

$$L_{h,k,l} = \left(0.94 \times \lambda \right) / \left(\beta \times \cos\theta \right) \quad (2)$$

where λ is X-rays wavelength; β is the full width at half maximum in radians; and θ is the Bragg angle.

2.6 Morphological structure

A scanning electron microscope (SEM) (Quanta 250 FEG, FEI, USA) with an accelerating voltage of 15 kV was used to investigate the microstructure and the surface morphology of the obtained cellulose microfibers.

2.7 Thermogravimetric Analysis (TGA)

In order to study the thermal stability of the extracted cellulose microfibers, thermogravimetric analysis (TGA) was performed using a Mettler Toledo TGA/DSC 3+ instrument. The scan was carried out from 25 to 600 °C at a heating rate of 10 °C/min and under nitrogen atmosphere.

3. Results and Discussion

3.1 Extraction method and cellulose yield

A stepwise totally chlorine-free procedure for the isolation of cellulose microfibers from retama raetam was proposed in this paper based on that adopted by Sun et al.[21]

Table 1. Comparison of the crystallinity index of cellulose microfibers from various sources.

Source	Crystallinity index (CrI)	Reference
Coconut palm leaf sheath	47.7%	Uma Maheswari et al.[22]
Sugarcane bagasse	50%	Jonjankiat et al.[23]
Agave fibers	64.4%	Reddy et al.[24]
Mengkuang leaves	69.5%	Sheltami et al.[3]
Soy hulls	69.6%	Alemdar and Sain[25]
Commercial microcrystalline cellulose	73.91%	Kale et al.[20]
Sisal fibers	75% ±1	Morán et al.[26]
Wheat straw	77.8%	Alemdar and Sain[25]
Retama Reatam	77.8%	This work
Hibiscus sabdariffa	78.95%	Sonia and Priya Dasan[27]
Palmyra palm Fruits	81.9%	Reddy et al.[28]

with modifications. These modifications involve increasing sodium hydroxide and hydrogen peroxide concentrations as well as improving treatment time and/or temperature to enhance non-cellulosic components removal. Therefore, the isolation of R. Raetam cellulose microfibers was successfully achieved without any additional harsh acid treatments (Table 1), which makes the suggested extraction process not only eco-friendly and cost-saving, but also yielding cellulose microfibers of higher crystallinity and smaller diameters as further confirmed by XRD and SEM results.

Cellulose microfibers yield was gravimetrically determined (calculated as the percentage of the extracted cellulose microfibers over the initial raw sample weight) and was found to be 52.1%. This yield value is higher than that reported in literature for cellulose microfibers extracted from Hibiscus sabdariffa fibers (38.6%)[27] and comparable to yield values of 52, 52 and 55% for cellulose microfibers extracted from Coconut palm leaf sheath[22], African Napier grass[29] and Palmyra palm fruits[28], respectively.

3.2 FT-IR spectroscopic analysis

Infrared spectroscopy is currently one of the most important analytical techniques available to scientists[30]. It presents a relatively easy method of obtaining direct information on chemical changes that occur during chemical treatments[31]. Furthermore, FT-IR analysis was conducted to investigate the presence of different functional groups in the isolated samples. Figure 1. shows the IR spectra of (a) untreated sample, (b) alkali treated sample and (c) bleached cellulose microfibers. As summarized in Table 2,

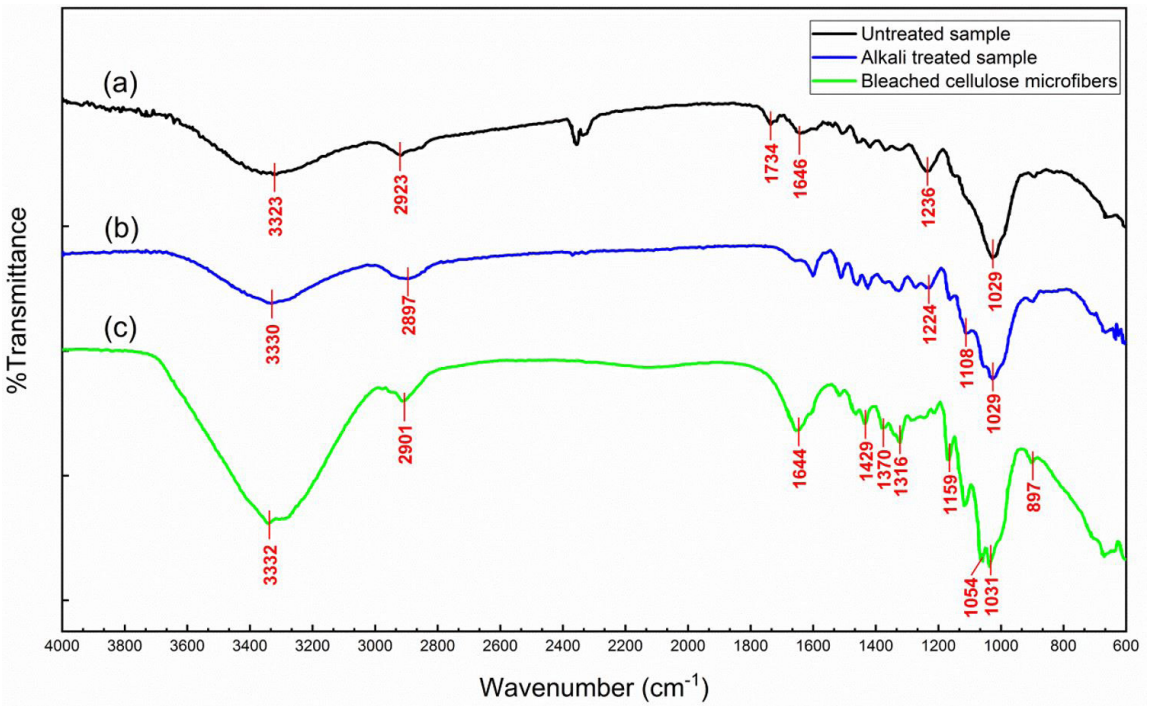

Figure 1. FT-IR spectra of (a) untreated sample, (b) alkali treated sample and (c) bleached cellulose microfibers.

Table 2. The main observed IR bands and their assignments.

Spectra	Wavenumber (cm^{-1})	Assignment	Ref.
(a), (b), (c)	876-897	C–O–C stretching at the b-(1→4)-glycosidic linkages	Oh et al.[32]
(a), (b), (c)	1029-1031	C–O–C pyranose ring skeletal vibration	Sun et al.[33]
(b), (c)	1051-1054	C–O–C pyranose ring skeletal vibration	Sun et al.[33]
(a), (b), (c)	1144-1159	C–O–C stretching at the b-(1→4)-glycosidic linkages	Oh et al.[32]
(a), (b)	1224-1236	–COO vibration of acetyl groups / C–O stretching of the aryl group	Reddy et al.[28]
(a), (b), (c)	1316-1327	C–C and C–O skeletal vibrations	Gao et al.[34]
(a), (b), (c)	1370-1374	O–H bending	Gao et al.[34]
(a), (b), (c)	1417-1429	CH2 bending	Gao et al.[34]
(a), (b), (c)	1644-1646	OH bending of the absorbed water	Alemdar and Sain[25]
(a)	1734	C=O stretching	Sain and Panthapulakkal[35]
(a), (b), (c)	2896-2922	C–H stretching	Kondo and Sawatari[36]
(a), (b), (c)	3323-3332	H-bonded OH groups stretching	Kondo and Sawatari[36]

the cellulose spectrum is similar to those reported in literature for cellulose fibers[37].

The band at 3332 cm^{-1} relates to the stretching of H-bonded OH groups, and the one at 2901 cm^{-1} to the C–H stretching[36], we observe that the band around 3332 cm^{-1} is narrower and has a higher intensity for cellulose, which demonstrated that the extracted cellulose contained more –OH groups than in untreated sample[10]. The band at 1644 cm^{-1} is associated with –OH bending of the absorbed water[25]. Typical bands assigned to cellulose at 1159 cm^{-1} and 897 cm^{-1} are due to C–O–C stretching at the b-(1→4)-glycosidic linkages[32]. The presence of these peaks showed the increase in the percentage of cellulosic components after the removal of non-cellulosic materials by chemical treatments[38]. The absorption peak at ~1734 cm^{-1} on the spectrum of the untreated sample (a) is attributed to the C=O stretching of the acetyl and uronic ester groups of polysaccharides[9,35], it is also related to the p-coumeric acids of lignin and/or hemicellulose[25], the absence of this peak after successive chemical treatments indicates the removal of most of lignin and hemicelluloses from the microfibers. Another indicator of lignin and hemicellulose removal during the chemical treatments is the significant decrease in the intensity of the peak around 1236 cm^{-1} which is related to the –COO vibration of acetyl groups in hemicellulose and/or the C–O stretching of the aryl group in lignin[28]. Noticeable peaks on spectrum (c) at 1429 cm^{-1} relates to the CH_2 bending and at 1370 cm^{-1} to the O–H bending. The absorbance at ~1316 cm^{-1} is attributed to the C–C and C–O skeletal vibrations[34]. The C–O–C pyranose ring skeletal vibration occurs in 1054 cm^{-1} and 1031 cm^{-1}[33].

3.3 X-ray Diffraction measurements

Figure 2 exhibits the XRD data of the extracted cellulose microfibers. The cellulose amorphous phase is characterized by the low diffracted intensity at a 2θ value of 19.12˚, whilst the peaks at 15.14˚, 16.25˚, 22.75˚ and 34.39˚ are attributable to the crystallographic planes of (1-10), (110), (200) and (004), respectively, which are characteristic of the typical cellulose I structure[29,39]. The crystallinity index (CrI) of the bleached microfibers was determined using Segal equation and was found to be 77.8%. Obviously, as illustrated in Table 1, this CrI value is higher than the value of 73.91% reported by Kale et al. for commercial microcrystalline cellulose from wood pulp[20]. Moreover, it is higher than the CrI values reported in the literature for cellulose microfibers isolated using different methods from various sources such as Sugarcane bagasse (50%)[23], Agave fibers (64.4%)[24], mengkuang leaves (69.5%)[3] and Soy hulls (69.6%)[25].

Furthermore, the crystallite size ($L_{h,k,l}$) which was found to be 3.62 nm is comparable to the reported sizes of cellulose crystallites (4 to 7 nm generally)[40]. However, the calculated $L_{h,k,l}$ value is much lower than that reported for hydrolyzed commercial microcrystalline cellulose (10.32 nm). The higher CrI and the lower $L_{h,k,l}$ values suggest that the adopted stepwise chlorine-free extraction treatments were effective in removing most of amorphous domains leading to break the bundles of cellulose fibers to form smaller cellulose crystallites[41,42].

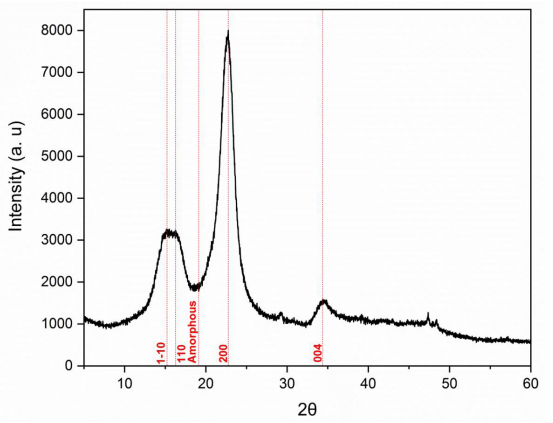

Figure 2. XRD diffractogram of the extracted R. Raetam cellulose microfibers.

3.4 Morphological properties of chemically purified cellulose microfibers

Figure 3 shows SEM micrographs of the chemical-purified cellulose microfibers. After they had been subjected to alkaline solution treatment and bleaching, the cellulose microfibers were separated into individual micro-sized fibers. These micro-sized cellulose fibers were reported to be composed of strong hydrogen bonding nanofibers[43]. The diameter of the microfibers is about 6-7 µm but the exact determination of their length is difficult.

As Table 3. demonstrates, the extracted R. Raetam cellulose microfibers are smaller in diameter as compared to those isolated by different extraction methods from various sources such as sisal fibers, agave fibers, coconut palm leaf sheath, soy hull and wheat straw[22,24-26]. Moreover, they are comparable to cotton and sugarcane bagasse microfibers extracted by sulfuric and nitric acid hydrolysis, respectively[23,44]. This morphology and smaller diameter would enable R. Raetam cellulose microfibers to be used for various applications ranging from reinforcing agents in biodegradable composites, to gel forming food and cosmetic additives[24,45].

3.5 Thermal stability

Investigating thermal properties of cellulose microfibers is a key factor for their applicability as reinforcing fillers in biocomposites[38]. Figure 4a and 4b shows, respectively, the thermogravimetric analysis (TGA), and the derivative thermogram (DTG) curves for both the raw and bleached samples. TGA curves show an initial weight loss below 155 °C, this initial drop (4.6% for raw sample and 6.5% for cellulose microfibers) was due to the evaporation of moisture bounded on the surface of the samples, chemisorbed water bounded inside the samples and/or the compounds of low molecular weight such as extractives presented in the raw sample[38,46], the presence of the absorbed water was affirmed previously through the FT-IR results. The main broader cellulose thermal degradation (50.84%) occurs over 179 °C and involves synchronous multi-processes such as dehydration, depolymerization and decomposition

Table 3. Comparison of the diameters of cellulose microfibers extracted from various sources by different extraction methods.

Source	Extraction method	diameter (μm)	Ref.
Sisal fibers	Alkali, peroxide and HNO_3/HAc treatments	12.8-31	Morán et al.[26]
Coconut palm leaf sheath	Chlorination, alkali and HNO_3/HAc treatments	10-15	Uma Maheswari et al.[22]
Soy hull	Alkali treatment and HCl Acid hydrolysis	10-15	Alemdar and Sain[25]
Wheat straw	Alkali treatment and HCl Acid hydrolysis	10-15	Alemdar and Sain[25]
Agave fibers	Chlorination, alkali and HNO_3/HAc treatments	8-14	Reddy et al.[24]
Hibiscus sabdariffa	Steam explosion and oxalic acid hydrolysis	10.04	Sonia and Priya Dasan[27]
Sisal fibers	Chlorination and alkali treatments	7-11.2	Morán et al.[26]
Cotton	Sulfuric acid hydrolysis	5-10	Chatterjee et al.[44]
Sugarcane bagasse	Nitric acid hydrolysis	5-10	Jonjankiat et al.[23]
Palmyra palm fruit	Chlorination, alkali and HNO_3/HAc treatments	4-11	Reddy et al.[28]
Napier grass fibers	Chlorination, alkali and HNO_3/HAc treatments	8.3	Reddy et al.[29]
Retama Raetam Stems	Alkali and alkaline peroxide treatments	6-7	This work
Jatropha Curcus L seed shell	Chlorination, alkali and HNO_3/HAc treatments	0.23-1.04	Puttaswamy et al.[45]

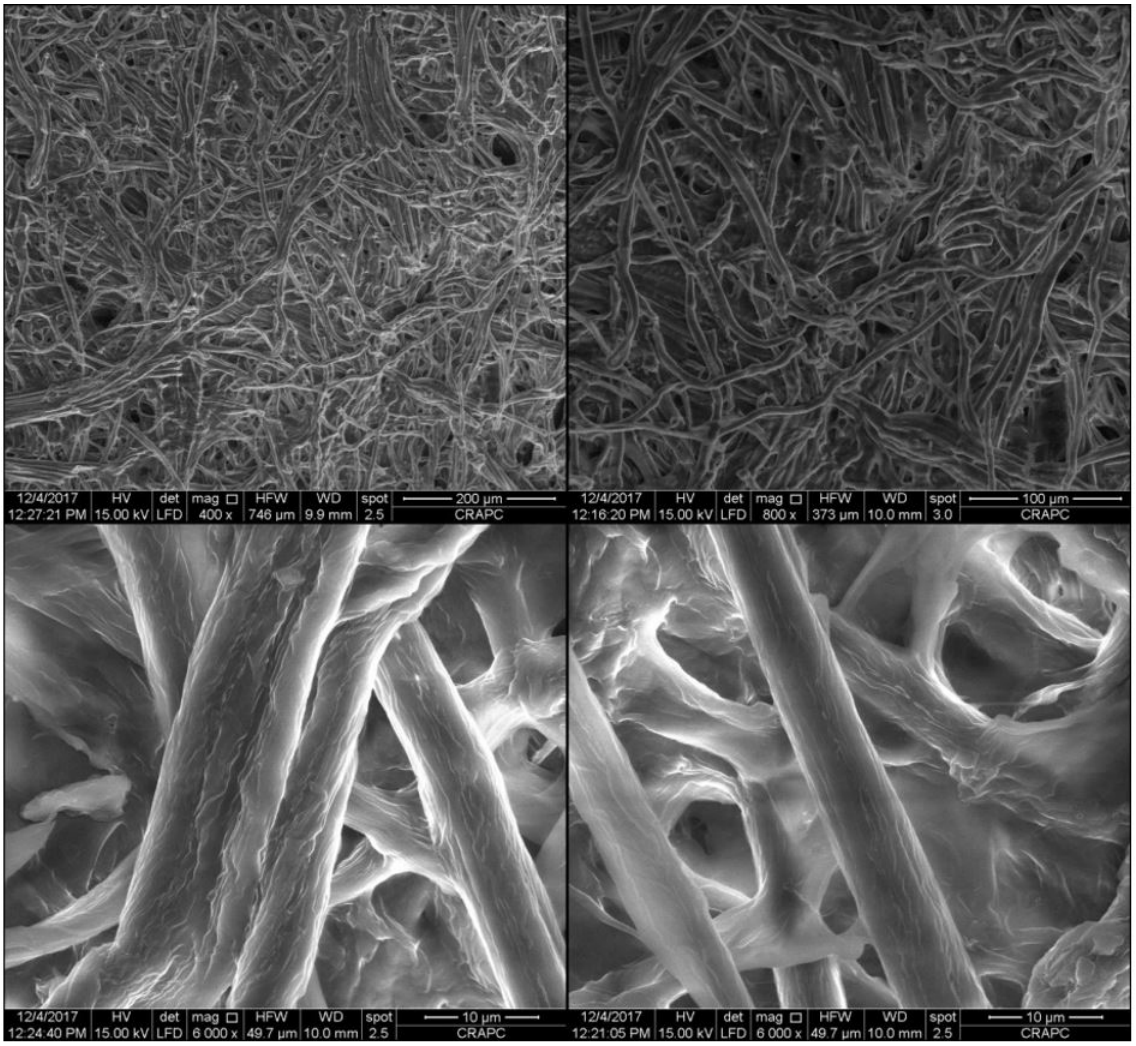

Figure 3. SEM images of the extracted R. Raetam cellulose microfibers.

of glycosidic units[47]. The raw sample showed separated pyrolysis processes within a wider temperature range, including thermal depolymerization of hemicellulose up to 273 °C, decomposition of cellulose up to 348 °C, and the degradation of lignin up to 536 °C in addition to its simultaneous decomposition with other degradation stages due to its complex structure[46]. DTG curves exhibited maximum decomposition rates at DTG_{max}=294 °C and

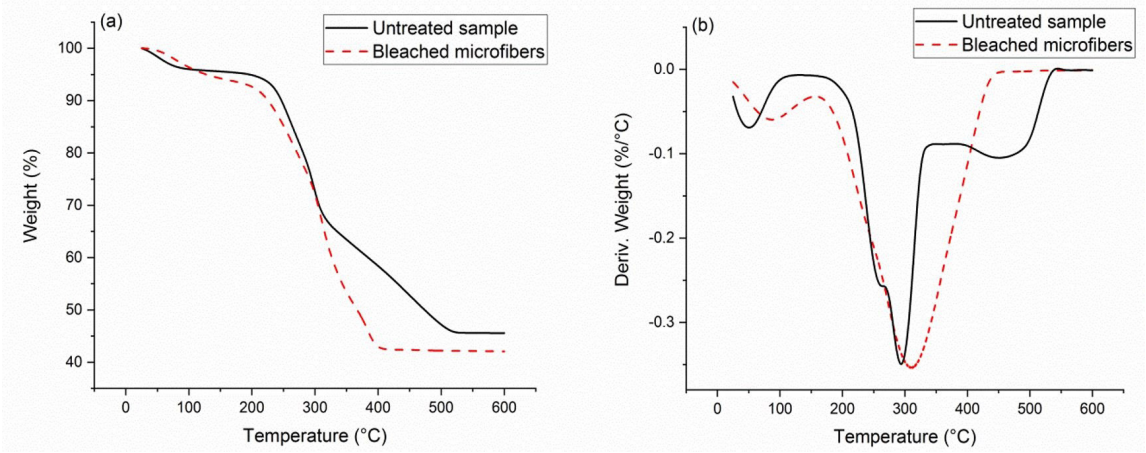

Figure 4. TGA (a) and DTG (b) curves of the untreated sample and the bleached cellulose microfibers.

Table 4. DTGmax and char yields of cellulose microfibers from different sources.

Samples	T_{max} (°C)	Char (%)	Reference
Date seeds cellulose microfibers	300	11	Nabili et al.[48]
Bamboo cellulose microfibers	328	13	Chen et al.[49]
Rice hulls microcrystalline cellulose	283	23	Adel et al.[50]
bean hulls microcrystalline cellulose	281	25	Adel et al.[50]
Onion skin cellulose microfibers	333	26	Reddy and Rhim[51]
Retama Raetam cellulose microfibers	311	42	This work
Cotton silver microcrystalline cellulose	340	57	Das et al.[52]
Jute microcrystalline cellulose	280	61	Das et al.[52]

311 °C for the raw and bleached samples, respectively. A shoulder can be clearly observed at 261 °C on the left side of the main peak of the raw sample DTG curve, which was due to initial decomposition of hemicellulose and non-cellulosic components[47], while the broadening at 245 °C on the microfibers DTG curve, could be an indicator of a broad distribution of molecular mass from cellulose or a residual content of hemicellulose which withstood the extracting procedures[26]. Finally, the formation of a charred residue took place (46% for raw sample and 42% for cellulose microfibers). The higher charred residue of the raw sample is due to the fact that the non-cellulosic components could induce higher char formation[38]. However, R. Raetam cellulose microfibers presented relatively high char yield when compared to literature (Table 4), indicating higher non-volatile carbonaceous material generated on pyrolysis[53] and could indicate also a good thermal stability of the extracted cellulose microfibers[20,54].

4. Conclusion

The main goal of this work was to investigate the viability of Retama Raetam as a novel, renewable and low-cost source of cellulose microfibers. The successful isolation of cellulose microfibers was achieved with a yield of 52.1% by stepwise chemical treatments. The FTIR results revealed that the chemical treatments removed most of lignin and hemicellulose from the sample. The extracted cellulose microfibers were highly crystalline native cellulose I, with a crystallinity of 77.8% and a crystallite size of 3.62 ṅm. The diameter of the micro-sized fibers was about 6-7 µm. TGA/DTG curves show a maximum decomposition peak at 311 °C and a high char yield. These findings proved that R. Raetam is a candidate renewable source for the production of cellulose microfibers and should stimulate further research on the use of these fibers for various applications such as cellulose nanocrystals preparation, reinforcement agent in green biocomposites and bio-fillers for polymer matrices.

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