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EXTRACTION OF CELLULOSE FIBERS FROM TÓ LEAF PETIOLES (*Calathea lutea*) AND CHARACTERIZATION

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Abstract

Tó leaf (*Calathea lutea*) is a widely distributed species in Tabasco, state of México. It could have a great potential for the production of natural cellulose fibers to be used as reinforcing material in bioplastic polymers. This study illustrates the extraction of cellulose from petioles of Tó leaves with Cazaurang modified method. The method consists of acid hydrolysis, chlorination, alkaline extraction and bleaching of the fibers. These chemical treatments permitted a yield of 26.25% of cellulose from petioles of Tó leaves. Fourier transformed infrared spectroscopy (FTIR) analysis revealed the efficient dissolution of the amorphous regions (lignin and hemicellulose) as a result of the applied methods. X-ray diffraction patterns indicated 38.09% crystallinity of the cellulose fibers and a crystal size of 2.6 Å, approximately. The obtained cellulose could be considered having a monoclinic form, corresponding to type I cellulose X-ray patterns, commonly found in plants.

Keywords: *Cellulose, Fibers, Calathea lutea, acid hydrolysis, FTIR, XRD.*

1. Introduction

Recently, as a result of the increasing concern for preserving the environment, special attention has been paid to the development of materials from natural and biodegradable sources. The biodegradability and environmental safety have become important in the design of new materials features [1]. According to Amigó et al. [2], vegetable fibers are those extracted from the plant kingdom such as seeds, stems, leaves, fruits and roots. They could be incorporated as reinforcing elements in thermoplastic biopolymers, improving mechanical properties of the compound and recovering losses from mechanical recycling of plastics.

Cellulose is the most abundant polymer in nature and it is primarily composed of the cell wall of plants, which gives support and rigidity to their structures [3]. It is frequently found in the vegetable kingdom in materials like wood, seeds, and agricultural waste, presenting fibrillary morphology. Arroyo et al. [1], defines cellulose as a linear chain of molecular rings of glucose ($C_6H_{10}O_5$) linked through a covalent bond C1 oxygen to glucose of a ring and the ring adjacent C4. Multiple cellulose chains are bound together by hydrogen bonds to form cellulose fibrils, in disordered regions (amorphous) or highly ordered (crystalline). Mainly, raw material for the production of cellulose comes from wood and cotton but due to the problems of deforestation it has had to resort to new sources [3]. Manufacturers uses the traditional synthetic fibers for reinforcement of thermoplastics (usually made of glass, carbon or aramid fibers), generating many environmental problems.

At present, the demand around the world of vegetable fibers is increasing. They are competing with wool, silk and synthetic fibers in qualities, strength, durability, color and gloss. They could be used in the health food industry and as a waste after being biodegradable [5].

The state of Tabasco (Mexico) owns a variety of tropical plant resources which are not yet studied in depth as a resource of cellulose fibers, and in this sense, they may have a susceptible economic value [5]. Tó leaf (*Calathea lutea*) is a highly abundant plant, easy to spread and grow. It is important to investigate its chemical nature and prove

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techniques extracting natural fibers from the regional Tabasco vegetation world to create possible new sources of pulp and fibers, applicable as biodegradable and environmentally friendly compounds.

2. Experimental

2.1 Material and methods

The biological material, petioles Tó leaves (*Calathea lutea*), was collected from the municipality of Huimanguillo, the region of Chontalpa, Tabasco state of México. Subsequently, it was sun dried and treated with an aqueous solution of 10% NaOH, with the aim of removing waxes, resins and pectins. The procedure is described below. Petioles Tó leaves were cut to a length of 20 cm, with four longitudinal cuts, and the solution of 10% NaOH penetrated perfectly into the samples: they were kept in this solution for 20 minutes when the boiling point was reached. After removing Tó leaves from the solution, they were allowed to cool and washed with running water, then manually shredded. The fibers were cut into 2 cm long and dried in an oven at a temperature of 60°C for 12 hours.

2.2 Cellulose isolation

Cellulose isolation was carried out using the pulping technique [6], consisting of four steps: (1) mild acid hydrolysis with 0.4% H_2SO_4 for one hour, and subsequent washing; (2) chlorination with 3.5% NaClO, stirring the solution in a water bath at 30°C to pH 9.2, washing with distilled water until neutral pH; (3) alkaline extraction with 20% NaOH under stirring for 1 h, followed by a washing process; (4) bleaching with a solution of 0.5% NaClO and continuous stirring for 1 hour, and a final wash to neutral pH. Then the material was manually shredded and placed in aluminum pan for one day at room temperature and then in an oven for 24 h at 60°C. The material was weighed, to determine the yield.

2.3 Instrumental analysis

Fourier Transform-Infra Red Spectroscopy (FTIR)

The chemical characterization of dried samples of petioles Tó leaves and extracted cellulose was carried out using the spectroscopy technique of Fourier transform infrared (FTIR) to determine the functional groups present in composition of the samples. Infrared spectrometer FTIR Nicolet Magna 460 Protegé in the absorbance mode, with a resolution of 4 cm-1 and 100 scans, was used. The tablets were made with 1 mg sample in 100 mg of KBr.

X-ray Diffraction (XRD)

The X-ray diffraction patterns, of petioles of Tó leaves samples and isolated cellulose, were analyzed with a Siemens D 5000 Diffractometer, using CuK radiation (λ = 1.5418 Å) at a 34 kV and a current of 25 mA, in a 2Θ angular range, between 10 and 30 degrees, making measurements every 0.04° for 6 s. The percentage of crystallinity of cellulose (Xc %) was calculated, according to Segal equation [7] :

$$Xc(\%) = \left(1 - \frac{I_1}{I_2}\right) X\ 100 \qquad (1)$$

where: I1 is the minimum peak intensity in the XRD patterns and I2 the crystalline peak intensity, respectively.

The crystal size (t) was calculated using the equation proposed by Scherrer [8]:

$$t = \frac{K\lambda}{B\cos\theta} \qquad (2)$$

where: λ is the wavelength of the radiation (λCu), B is the width at half height of the diffraction peak of the sample, and K is the dimension less form factor which has a typical value of about 0.9, but varies with the actual shape of the crystallites.

Scanning Electron Microscope (SEM-EDX)

Scanning electron microscope, JEOL JSM 610 LA model, was used to observe the surface morphology of the isolated cellulose fibers from petioles leaves of Tó. The effect of the chemical treatments was assessed using a comparison of the untreated samples. EDX analysis revealed the main elements as a part of the cellulose after its extraction of Tó leaves.

3. Results and discussions

3.1Fiber yield

The extracted cellulose fibers, from petioles of Tó leaves, showed an average of 26.25 ± 1.6% of yield, as final dried product.

3.2 Fourier transform-infra red spectroscopy (FTIR) analysis

The FTIR spectrum of untreated samples of petioles leaves of Tó is shown in Figure 1. The peaks at certain frequencies, characteristics for different functional groups are: 3435 cm^{-1} corresponding to stretching vibrational

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alcohols (OH) [9, 10] and 2920 cm^{-1} for stretching OH methyl and methylene groups [10, 11]; stretching vibrational peak of C=O at 1732 cm^{-1} is attributed to waxes and pectins [11, 12]. The peak at 634 cm^{-1} is due to the bending mode vibration of the absorbed water, with some contributions from carboxylate groups [9].

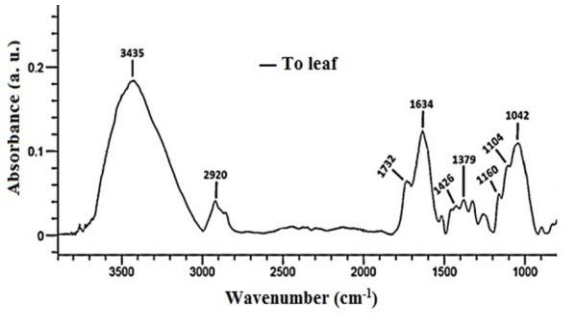

Fig: 1. FTIR spectrum of petioles of Tó leaves (*Calathea lutea*).

The FTIR spectrum of extracted cellulose from petioles of Tó leaves is shown in Figure 2. The peaks at certain frequencies, characteristic for different functional groups are: the peak at 1732 cm-1, attributed to hemicelluloses, pectin and waxes [11, 12], which intensity is lower of that for untreated Tó leaves (Fig.1), confirming the effective removal of these compounds after the chemical treatment; the peak at 1050 cm^{-1} is due to the ether linkage from lignin or hemicellulose. This last peak indicates that traces of lignin and hemicellulose are still present. The peak at 997 cm^{-1} is attributed to the β-glycosidic linkages of glucose ring of cellulose, probably as a result of the acid treatment of the leaves. The characteristic peaks shown in FTIR spectrum are in a good agreement with the reported [13].

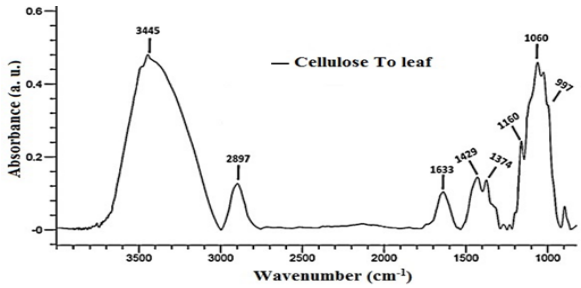

Fig: 2. FTIR spectrum of extracted cellulose from petioles of Tó leaves (*Calathea lutea*).

3.3 X-ray diffraction (XRD) analysis

The diffraction patterns, of untreated samples of petioles of Tó leaves and those of extracted cellulose, are shown in Figure 3 and Figure 4, respectively. It can be observed that Tó leaf cellulose has a well pronounced crystalline structure, due to hydrogen bonding and van der Waals interactions, existing between adjacent cellulose molecule, compared to hemicellulose and lignin, which are amorphous in nature. The peak near 2θ = 21.8º, observed in both X-ray patterns, is characteristic of the cellulose [14, 3], and peaks at 2θ = 12.1°, which are 21.4º and 34.6º correspond to the structure of cellulose I, commonly found in the natural vegetable fibers [15, 16, 17](Visakh et al, 2010; Jin and Kamdem, 2009 and Isogai et al., 1989).

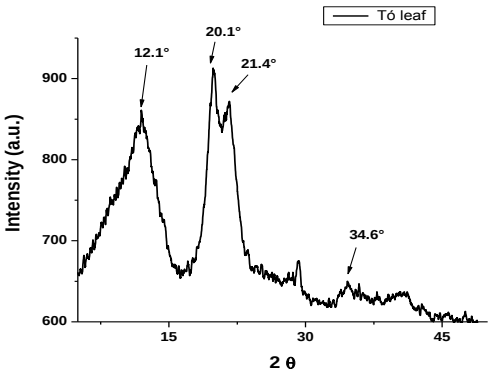

Fig: 3. X-ray diffraction patterns of petioles of Tó leaves (*Calathea lutea*).

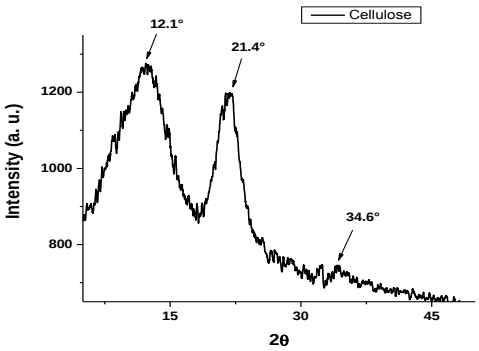

Fig: 4. X-ray diffraction patterns of extracted cellulose fibers after the chemical treatment of petioles of Tó leaves (*Calathea lutea*).

It can be seen (Fig. 4) that the intensities of the cellulose peaks of fibers, obtained after the acid treatment of petioles of Tó leaves, are higher, compared to those in Figure 3. This fact indicates the efficient removal of noncellulosic polysaccharides and dissolution of amorphous zones [18] (Chen and Yokochi., 2000), after

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the chemical treatment of the fibers. As a result, their crystallinity index increased (Table 1) and the crystal size was decreased, compared to those of untreated Tó leaves.

Table 1: Crystallinity index and crystal size of petioles of Tó leaves (*Calathea lutea*), and of extracted cellulose fibers.

Samples	Petioles of Tó leaves	cellulose fibers
Crystallinity index (%)	22.60	38.09
crystal size (nm)	2.96	2.60

It is reported that the crystal size of cellulose may be increased or decreased by the effect of several factors as the source of the cellulose, method of its extraction and after pumping treatments [8, 19]. According to reported results, the obtained cellulose in this study could be considered having a monoclinic form, corresponding to type I cellulose X-ray patterns, commonly found in plant tissues [1], specifically cellulose-1β stable monoclinic crystals.

3.4 Scanning electron microscopy (SEM-EDX) analysis

Based on SEM images, the cellulose fiber diameter average was determined and this was approximately 1.71 ± 3.9061µc for the cellulose extracted from petioles of Tó leaves (Fig.5).

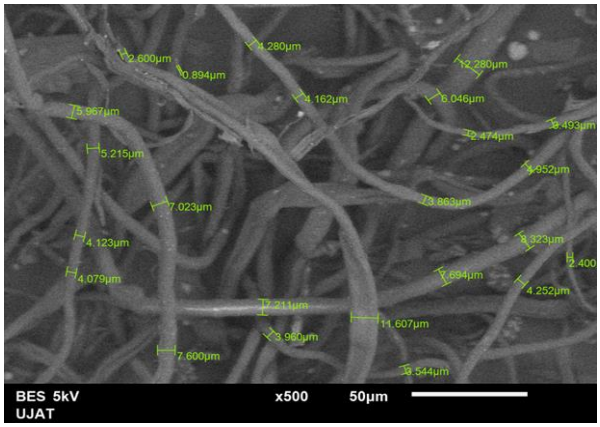

Fig: 5. Micrograph (SEM) showing the diameter of the cellulose fibers extracted from petioles of Tó leaves.

Elemental analysis SEM-EDX revealed that the extracted cellulose fibers from Tó leaves contain as main chemical elements carbon (C) and oxygen (O). Traces of chlorine (Cl), residues of bleaching and chlorination steps were detected, and calcium (Ca) probably originated from the washing water.

4. Conclusions

Cellulose fibers were extracted from petioles of Tó leaves (*Calathea lutea*), after acid chemical treatment and several steps using Cazaurang modified method. The procedures permitted a yield of 26.25% of cellulose. Patterns of X-ray diffraction of the extracted cellulose showed an increased intensity, suggesting a higher crystallinity than that of the Tó leaves, probably due to the efficient dissolution of the amorphous regions by the acid treatments. The infrared analysis (FTIR) confirmed these results and the increase of the crystallinity of the extracted cellulose was associated with the disappearance of the typical bands of the removed hemicelluloses and lignin. The calculated crystallinity of the extracted cellulose fibers was 38.09% and their crystal size of 2.6 Å, approximately. The obtained cellulose could be considered having a monoclinic form, corresponding to type I cellulose X-ray patterns, commonly found in plant tissues, specifically cellulose-1β stable monoclinic crystals. The obtained results could be considered promising for the use of these natural fibers as reinforcement in biodegradable materials and paper production among other uses.

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