

Bioplastic composed of starch and micro-cellulose from waste mango: mechanical properties and biodegradation

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Abstract

Waste mango was used to obtain starch and micro-cellulose for the production of bioplastic. Three different formulations were made: positive control or cotyledon starch/glycerol; SC₁ or cotyledon starch/glycerol and cellulose at 0.1% and SC₅ or cotyledon starch/glycerol and cellulose at 0.5% w/w. The bioplastics were mechanically analyzed (tensile strength, elongation and Young's modulus) and, aerobic biodegradation analysis was realized with a standard test method based on the amount of material carbon converted to CO₂. The mechanical tests indicated that with the addition of cellulose, the bioplastics improved their mechanical properties. The biodegradation at 30 days showed 93 and 94% for SC₁ and SC₅. Therefore, the biodegradation of bioplastics depends on both, the addition of cellulose and the environment where they are placed (e.g., soil characteristics: pH level, C:N ratio, moisture). These bioplastics offer new opportunities for fast degrading biomaterials in agricultural applications (padding and protection bags).

Keywords: *bioplastic, cellulose, cotyledon starch, biodegradation, mechanical tests.*

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1. Introduction

Since the appearance of synthetic plastics in the past century from the refining of petroleum, synthetic plastics have become the most commonly used materials in different industrial sectors, mainly due to their low cost, lightweight and durable materials with adequate mechanical and chemical properties^[1,2]. The heterogeneity of synthetic polymers as well as the variability in their properties have allowed for their use in generating a wide range of products that have several benefits in the medical and technological sectors^[2,3]. However, pollution from plastic waste has developed to become a great threat to ecosystems, especially aquifers with the oceans been the most affected^[4,5]. An estimated 8.3 billion metric tons of plastic are produced worldwide, and only 9% of used plastic has been recycled, 12% are incinerated and 79% accumulated in landfills or natural environment as litter^[6,7]. In recent years, interest in the development and applications of degradable plastic (i.e., plastics produced from fossil materials) or bio-based plastics and fillers (i.e., plastics synthesized from biomass or polymers from renewable resources) with functionalities and processabilities comparable to traditional petrochemical-based plastic has attracted increasing attention^[8].

Biodegradable polymers based on natural polysaccharides, such as starch and cellulose, are used as raw material to develop biodegradable films since they have the capability

of forming a continuous matrix and they are a renewable and abundant resource^[9-12]. Starch is one of the most promising natural polymers because of its inherent biodegradability, overwhelming abundance and annual renewability^[12-14]. In addition to its status as a renewable resource, starches offer very attractive features: its low energy consumption involved in its production, its potential to add value to by-products from other industries, its characteristic biodegradability, among others^[14,15]. Starch is essentially composed of a mixture of polysaccharides: amylose, amylopectin and a minor fraction (1% to 2%) of non-glycosidic conformation^[16]. Amylose is avowed as linear polymer (poly- α -1,4-D-glucopyranoside) mainly distributed in the starch granule amorphous part and, a branched polymer amylopectin (poly- α -1,4-D-glucopyranoside and α -1,6-D-glucopyranoside), that gives place to the crystalline domains^[17]. In many native or normal starches, around 70 – 80% of the mass of starch granule contains amylopectin and about 20 - 30% amylose^[16,17]. The studies of Mali et al.^[18] and Bae et al.^[19] reported that starches with high amylose content (~30%) have great film-forming properties in comparison with other starches with lower amylose content. Taking into consideration mango starch which can be used as raw material for the manufacture of biodegradable films, since the concentration of mango starch amylose is high enough (31.4%)^[20], and is assumed

to be the basis for good biodegradable films. In this sense, it is important to point out that in Mexico the mango is one of the fruit crops with the highest production, reporting in 2019 a production of 2 089 000 tons^[21], however, the low opening in the national and international markets have generated an increase in the generation of post-harvest waste. The waste mango currently it is not widely used and which represents an environmental problem due to the followings: lack of distribution channels of mango, inadequate management of the large volumes of residues that are generated, and environmental policies that regulate the deposition of post-harvest mango residues. Therefore, this waste mango can be used as raw material to obtain starch to develop bioplastic film.

Biodegradable plastic can be degraded by naturally occurring microorganisms such as bacteria, fungi, and algae to yield water (H₂O), carbon dioxide (CO₂) and/or methane (CH₄), biomass, and inorganic compounds^[22,23]. Therefore, the study of the biodegradability of biodegradable plastic plays a significant role in the development of biopolymers. For a bioplastic to be considered biodegradable, there must be a significant change in the chemical structure and also 90% of the substance is converted to carbon dioxide in less than six months^[24].

As for cellulose, this polysaccharide is considered the most abundant natural polymer in the world with the characteristic of being renewable and biodegradable. In the same way it has also been a classic example of a biopolymer used as matrix^[25,26]. Cellulose is commonly found in the primary cell wall of green plants; however, it is also produced by some bacteria^[27]. It is completely a linear polymer whose basic unit is *D*-glucose which is successively linked through a glycosidic bond in configuration β (1-4) to form cellobiose molecules. Cellulose chains are arranged in microfibrils of polysaccharides which aids in the stability of plant structures which also suggests that cellulose is a biomaterial with high strength and other superior mechanical properties^[23,24]. However, the native fibers of cellulose are rigid mainly due to the components that form them (Lignin, cellulose and hemicellulose), being lignin, the only component made up by aromatic organic units which gives it a high rigidity that allows it act as adhesive of the other two. Therefore, it became necessary to carry out series of chemical and thermal treatments to remove them from the fibers without damaging the structure of the cellulose microfibrils^[28].

For all of the above, many researchers have seen the development of biodegradable plastic as a promising solution in reducing the negative influence of plastic waste on the environment since they are environmentally-friendly and are also made from renewable resources and not from fossil fuels.

Regarding these concepts, the work aimed to evaluate the effect of the incorporation of micro-cellulose in the mechanical properties and biodegradation in soil of bioplastics prepared from cotyledon starch/glycerol/micro-cellulose.

2. Materials and Methods

2.1 Materials

Ataulfo mango waste (*Mangifera caesia* Jack ex Wall) were supplied by a regional mango producer in El Arenal,

Guerrero, Mexico. Sodium hydroxide, sulfuric acid, hydrogen peroxide, citric acid, ethanol, sodium chlorite, hydrochloric acid, glycerol, phenolphthalein and distilled water were supplied by Merck-Millipore™. Benzene, acetylacetone, dioxane and potassium hydroxide were purchased from Sigma-Aldrich™. All chemicals were used as supplied by the manufacturer.

2.2 Starch extraction

The mango cotyledons were cut into 2 cm pieces and placed in a container kept in a continuous flow oven for 24 h at 40 °C. The cotyledons were ground for 5 min using a semi-industrial mixer with distilled water and citric acid 3% (w/v) solution^[29]. The material obtained was passed through 60, 80 and 100 mesh sieves respectively, and the resulting mixture was centrifuged at 7000 x g for 5 min at 4 °C. Finally, the starch obtained was kept in a continuous airflow oven at 40 °C for 24 h, pulverized in a mortar, passed through 100 mesh sieve and stored in double click bags^[30].

2.2.1 Amylose content

The apparent amylose content was determined according to the method suggested by Gilbert and Spragg^[31], potato starch was used as standard. 0.1 g dry starch sample was weighed and 1 mL ethanol (95%) was added followed by 9 mL of 1 M NaOH solution. The sample was kept for 10 min in a shaking water bath at 85 ± 0.2 °C. The starch solution was cooled and transferred into a volumetric flask and the volume made up to 100 mL mark with distilled water. 2.5 mL of starch solution was taken into 50 mL standard flask; 0.5 mL of 1 M acetic acid was added followed by 1 mL of stock iodine (0.2 g I₂ / 2.0 g KI/ 100 mL) and the solution made up to the 50 mL mark with distilled water. A sample was taken from this solution and transferred to a Perkin-Elmer Lambda 3B double beam UV/ visible spectrophotometer to obtain the absorbance of the sample at a wavelength of 600 nm. The blue value was calculated using the formula^[31]:

$$\text{Blue value} = \frac{\text{Absorbance at } 620 \text{ nm} \times 4}{\text{Concentration} \left(\frac{\text{mg}}{\text{dl}} \right)} \quad (1)$$

2.3 Extraction of micro-cellulose fibers

The fibrous endocarp was cut and sieved with a 100-mesh sieve. The bagasse was washed with distilled water for 20 min with stirring and at room temperature. It was filtered using filter paper, and the samples were dried in a continuous flow oven at 40 °C for 24 h^[32].

2.3.1 Alkaline and bleached treatment

The dried fibrous endocarp was subjected to alkaline treatment^[30] using a 2% (w/v) sodium hydroxide solution in a ratio of 1:20 (fiber: solution), keeping it stirred for 2 hours at 80 °C (Figure 1a). The samples were oven-dried in continuous flow oven at 40 °C for 24 h, and ground using an E3303.00 mini cutting mill (Eberbach Corp.) The fibers were bleached (Figure 1a) in a 1:20 ratio (fiber: solution) in a solution composed of H₂O₂ (v/v) and 4% NaOH (w/v),

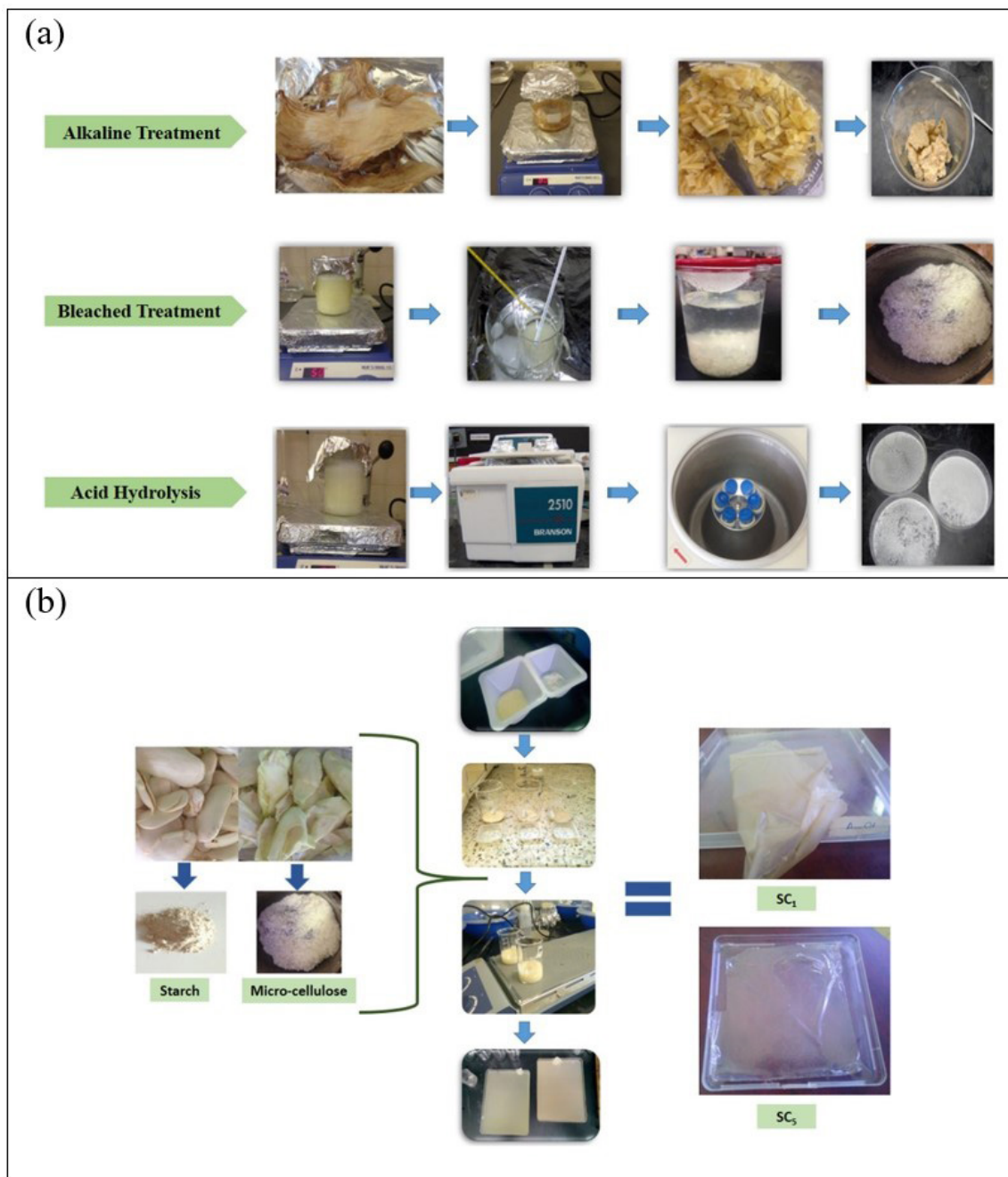


Figure 1. Illustrate kind steps of: (a) chemical (alkaline, bleached and acid hydrolysis) and thermal treatments (sonication) applied to cellulose to obtain micro-cellulose and (b) bioplastic preparation (from cotyledon starch and micro-cellulose).

stirred for 2 h at 50 °C and washed with distilled water and dried at 40 °C for 24 h^[33].

2.3.2. Acid hydrolysis

Cellulose fibers were prepared by acid hydrolysis (Figure 1a), using the method proposed by Cordeiro et al.^[30] with some adaptations. Acid hydrolysis was carried out using H₂SO₄ solutions at 52% (w/w) in a 1:20 ratio (fiber: solution), with hydrolysis time of 2 h at 45 °C. The sample was centrifuged at 6300 x g for 10 min at 10 °C and, the

resulting suspension was sonicated for 5 min at a power of 99 W using a Branson™ 2510MT ultrasonic cleaner, according to the modified techniques of Szymańska-Chargot et al.^[34] and Kasuga et al.^[35].

2.3.3 Chemical analyses

The lignin content was determined using ethanol-benzene solvent for 5 h^[36]. The mixture was washed with distilled water and dried in an oven at 103 °C for 1 h, then treated with 72% H₂SO₄ for 2 h while stirring at 37 °C.

The material was diluted to 3% H₂SO₄ and stirred at 80 °C for 4 h; finally, the sample was dried at 105 °C for 1 h and placed in a desiccator for cooling until a constant weight was obtained. The lignin content was calculated using Equation 1, where: *A* is the weight of lignin and *W* is the oven-dry weight of the test specimen^[37].

$$\text{Lignin \%} = \frac{A100}{W} \quad (2)$$

Holocellulose was measured by treating dry fibers with an acidified aqueous sodium chlorite solution in an acid medium at 75 °C for 1 h^[38] until the fibres were bleached. The determination of cellulose in fiber was carried out employing a process with aqueous solutions of acetylacetone, dioxane, and hydrochloric acid^[39]. The hemicellulose content was theoretically calculated from the difference in the holocellulose and cellulose contents.

2.4 Bioplastic preparation

Bioplastic were prepared from cotyledon starch and micro-cellulose using the method described by Pranoto et al.^[40] (Figure 1b). The solution was prepared from cotyledon starch (4% starch, w/v) with the addition of micro-cellulose (0.1 and 0.5%, w/v) and glycerol (0.9%, w/v) used as a plasticizer. The solution was prepared in a beaker with 95 mL distilled water at ambient temperature, then the cotyledon starch was added and stirred for 10 min at 30 °C. Thereafter, the solutions were kept at 60 °C for 10 min. Glycerol and micro-cellulose were then added and the solutions were made up to 100 mL with distilled water and heating was applied until it reaches 75 °C. The filmogenic solution was cast onto the glass plates (30 cm x 20 cm) and dried at 40 °C for 12 h in a continuous airflow oven (Barnstead International™, Model Imperial V). The films obtained were peeled off and stored in a desiccator at 25 °C with relative humidity (RH) of 57% provided by a saturated solution of NaBr.

2.4.1 Biodegradation test

The test was carried out based on the ASTM D5988-12 standard^[41]; which establishes the test method to determine the degree and rate of aerobic biodegradation of materials in contact with soil. The soil had the following characteristics: pH 8.1, carbon 9.64%, nitrogen 0.19%, humidity 6.80%, C:N ratio (16:1). The desiccator was conditioned with layers of soil on the bottom on which were placed the bioplastics and polymer plastic. Inside the desiccator, 20 mL of 0.5 N potassium hydroxide (KOH) solution were placed in a 100 mL beaker, as well as another beaker with 50 mL of distilled water. The desiccator was sealed, and placed in a dark place at 21 ± 2 °C with a humidity of 50%. The carbon dioxide (CO₂) produced was trapped by the KOH solution. In addition, a positive control desiccator was included; no biodegradable film sample. Periodically, the amount of CO₂ produced by the microorganisms present in the soil was determined. The CO₂ released from the system was fixed in the 0.5 N KOH solution and titrated with 0.25 N HCl, using phenolphthalein as an indicator. The percentage of biodegradation was calculated according to the following equation^[41]:

$$\% \text{ Biodegradation} = \frac{\text{mg of CO}_2 \text{ produced}}{\text{mg of theoretical CO}_2} \times 100 \quad (3)$$

2.4.2 Weight loss from samples

The samples were cut obtaining an area of 4 cm² and placed on the surface of the Petri dish containing the soil. The weight loss evaluation was determined gravimetrically, the samples were weighed before and after 5, 10, 15, 20 and 30 days of degradation. A polymer plastic bag (Low-density polyethylene, LDPE) was used as a reference^[42]. The samples were carefully cleaned, then washed with distilled water, until all traces of soil were removed. Then, the samples were dried for 24 h at 60 °C. Finally, the films were weighed and the final weight was recorded. The calculation of the loss in weight was carried out using Equation 4 and photographic monitoring and analysis by SEM were carried out.

$$\% \text{ weight loss} = \frac{\text{starting weight} - \text{final weight}}{\text{starting weight}} \times 100 \quad (4)$$

2.5 Structural characterization

To detect changes in the structure of fibers with alkaline treatment as well as chemical and structural analysis of starch, Fourier-transform infrared (FT-IR) spectra were performed on a Perkin Elmer Spectrum (100/100 N model, Shelton, CT, USA) in the range of 4000-650 cm⁻¹ in the transmittance mode, with a resolution of 16 cm⁻¹ and 8 scans^[43]. Scanning electron microscopy (Carl Zeiss EVO LS 10) was used to observe the morphology of the samples at an accelerating voltage of 25 kV, with a resolution of 3-10 nm; spot size of 2 was used to image the samples^[44].

2.6 Mechanical properties

The mechanical properties of the bioplastics were determined using TAXT2i texturing equipment (Stable Micro Systems™, Surrey, UK), equipped with a 25 Kg load cell. The bioplastics were studied following the ASTM method D-882-02^[45]; the bioplastic samples were cut into rectangles that were 10.0 cm long and 1.0 cm wide. The tensile strength (TS), elongation (%E) and Young's modulus, were determined using a deformation rate of 1 mm/min.

3. Results and Discussions

3.1 Raw material

3.1.1 Starch, amylose and amylopectin content

Cotyledon starch granules presented an oval and elliptical morphology, a size from 2 to 17 μm, with an average of 9.2 μm. This size is similar to that reported by cotyledon starch of five mango varieties grown in India, where the granules presented intervals of 1.5 to 28 μm in diameter^[46]. The resulting starch composition showed a significant amylose content of 29.19%; nevertheless, it was lower than the amylose content of 30.45% and 32.0% found in seeds from Tommy Atkins an Alphonso varieties mango^[47,48]. Morrison and Azudin^[49] and Gao et al.^[50] have reported that the amylose content may vary due to environmental effects and cultural conditions. In general, it has been reported that

the amylose content present in native starches is between the values of 18 and 30% and 70 to 82% for amylopectin content, in addition to the presence of other constituents such as lipids, proteins and minerals^[51,52]. The lower the amount of these other constituents and the higher the amount of amylose, the better the film and coating formation as amylose is directly linked to the chemical and physical characteristics of the film^[53].

3.1.2 Chemical analyses of the fibers

On the other hand, since what is sought in a fiber with hydrophobic properties, that can help and avoid deterioration due to high humidity conditions, it is necessary to perform a chemical and thermal treatment, that can help to remove glue from the fibers without damaging the structure of the micro-cellulose fibers. After chemical and thermal treatment the fibers showed a uniform surface and rectangular shape with sizes ranging from 40 to 400 μm in length, in comparison with the values of 200 to 250 μm that presents the native fiber (without chemical and thermal treatment). Likewise, the application of the fibers is desired for reinforcement in polymer blends, it is desirable to remove amorphous constituents (hemicellulose and lignin) and increase the content of cellulose, which is the crystalline phase of the fiber^[32,34]. The decrease in contents of lignin and hemicelluloses was recorded for sample modified in NaOH and H_2SO_4 solutions. The results of lignin were 10.03 wt % for native fiber and 0.72 wt % for fiber subjected to a modified acid-alkaline treatment; this result showed that the lignin determined by the Klason method, was partially removed in cellulose fibers. The calculated hemicellulose decreased from 28.63 wt % to 1.49 wt % for native and modified fiber, respectively. Based on the results, the lignin content from this study (10.03 wt %) is close to the value of 9.0 wt % reported by Guzmán et al.^[53]. However, different values are obtained given that the food-processing industry method was employed which is similar to the Klason lignin extraction process, with the variant that a final step of 1 h calcination at 550 °C is done. A major difference was observed in the lignin results in relation to the values of 5.76 wt % and 6.97 wt % of lignin in mango peel and mango by-products, respectively^[54,55]. On the other hand, the hemicellulose content of 29.75 wt % obtained in this study was lower than those of mango peel (32.5 wt %) and mango by-products (31.75 wt %)^[53,56]. A possible explanation is the fact that hemicellulose was not completely removed by the neutral detergent solution used for determining the Neutral Detergent Fibre in the food-processing industry method, as such this method was not used for this study. In addition, the acid concentration (52% w/w) used in the hydrolysis process allowed the destruction of both the amorphous and crystalline regions of cellulose and the decrease in contents of lignin and hemicelluloses^[57-59].

3.1.3 Fourier transform infrared spectroscopy (FTIR)

Through acid-alkaline treatments, hemicellulose and lignin were removed, as this also improves the thermal stability and tensile strength of the fibers, an essential property necessary for the elaboration of bioplastics. Typical bands assigned to cellulose were observed in the region of 1745 - 900 cm^{-1} , where the signal at 1635 cm^{-1} correspond to vibration of water

molecules absorbed in cellulose and starch. The absorption bands at 1420, 1366, 1334, 1027 cm^{-1} and 896 cm^{-1} belong to stretching and bending vibrations of CH_2 and CH, OH and C-O bonds in cellulose. The native and micro cellulose fibre infrared spectrum subjected to chemical treatment showed a peak at 894 cm^{-1} (Figure 2), signal was found in the polysaccharide absorbing region (950-700 cm^{-1}) which is associated with the anomeric carbon present in cellulose and which also reveals the component structures of β -glucans^[60]. The stretching of C-O-C bonds of the alkyl-aryl ether (at 1232 cm^{-1}), a compound that belongs to the structure of lignin can be observed only in the native sample since this signal disappears after chemical treatments confirming lignin removal. The 1745 cm^{-1} peak corresponds to the stretching of the C=O groups linked to aliphatic carboxylic acid and ketone and the residue of hemicellulose or the ester links of the carboxyl group in the ferulic and p-coumaric acids of lignin and hemicellulose^[61,62]. This signal decreased when the sample is subjected to acid treatment, which made it possible to verify the removal of lignin and hemicellulose. The bands within 2922-2854 cm^{-1} are ascribed to the aliphatic materials such as cutin, waxes and cutan present in hemicellulose and lignin^[63].

The band at 2876 cm^{-1} is attributed to CH stretching vibration of all hydrocarbon constituent in polysaccharides. The FTIR spectra of the fibre samples under study provided evidence that the fibers subjected to a chemical treatment has featured the removal of lignin and hemicellulose. A significant decrease is noted in the infrared spectrum when the sample was subjected to an acid-base treatment due to the removal of these components^[64].

In the case of native cotyledon starch, the FTIR spectra provided information about the presence or absence of specific functional groups and can give an even deeper insight into the granule starch structure. In the region within the spectrum of 1700 - 800 cm^{-1} the fingerprint was found, the region where the characteristic peaks in polysaccharides occur. In the region between 3000-2800 cm^{-1} the C-H stretching was found and finally the region between 3600-3000 cm^{-1} the O-H stretch region was found. The band in 930 cm^{-1} is attributed to the α -1,4 glycosidic bonds, (C-O-C) in starches. The peak at 1163 cm^{-1} is attributed to C-O coupling modes and C-C stretching of the polysaccharide structure^[65]. A signal was observed at 2920 cm^{-1} which is related to the amount of amylose and amylopectin present in the starch granules. A peak appears at 2850 cm^{-1} which is related to the alkyl

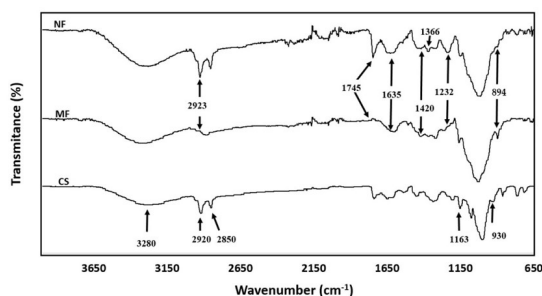


Figure 2. FTIR spectra of cotyledon starch (CS), native fibre (NF) and modified fibre (MF) subjected to a chemical treatment.

and aliphatic groups (H-C-H) present in hemicelluloses and lignin^[64]. The presence of this peak is attributed to a thin filament composed of fiber that covers the cotyledon. The maximum absorption was found at 3280 cm⁻¹, which is considered due to the vibrational tension stresses associated with the free, intermolecular and intramolecular bonds of the hydroxyl groups of starch^[66]. Infrared spectroscopy showed direct information on chemical changes that occur during chemical and thermal treatments applied to cellulose, in the same way FTIR allowed to observe the presence of different functional groups in the isolated samples (e. g. starch).

3.2 Bioplastics

3.2.1 Mechanical properties

Mechanical properties of starch/micro-cellulose-based bioplastics and conventional plastic (low-density polyethylene, LDPE) are shown in Table 1.

The tensile strength (TS), Elongation (%E) and Young's modulus of the starch bioplastic sample (positive control) was lower when compared with other samples. However, samples with cellulose presented higher values. These results suggest that at higher cellulose content, mechanical properties of the material tends to increase, since the interaction between the reinforcing material and the polymeric matrix determines the mechanical properties, in terms of stress, elongation and Young's modulus^[67].

The use of cellulose in the preparation of the bioplastic makes them more flexible and elastic compared to the positive control sample made of starch without cellulose, thus its mechanical properties were improved. According to Müller et al.^[68], the addition of 3 and 5% cellulose fibers from various sources considerably increases the tensile strength and reduces its elongation capacity. These results agree with the findings of Prachayawarakorn et al.^[69] in a matrix of starch extracted from rice and reinforced with 5 and 10% cotton cellulose, and also with the findings of Sudharsan et al.^[70], in a matrix of tamarind seed starch and 4% sugarcane bagasse cellulose, where TS increased due to the addition of cellulose. On the other hand, with regard to the polymer plastic which exhibits a typical behavior of a flexible polymer due to its excellent mechanical resistance, with a TS value of 12.582 MPa, close to that presented by other studies^[71,72] in relation to the force supported before breaking. The value of %E was higher, 41.492 MPa; the higher this value, the more the material will be able to stretch before breaking. Nevertheless, SC₁ and SC₅ still exhibited good strengths and flexibilities compared to the polymer plastic, which is attributed to the increased concentrations of cellulose. Cellulose acts as a reinforcing material which causes the value of the tensile strength of bioplastics to increase and

making it have a behavior so close to conventional plastic. This, in addition to the amylose content (29.19%) found in this type of starch, with a higher amylose content could have bigger crystalline regions have lower tensile strength and Young's modulus, and higher elongation at break because of their greater mobility^[73-75]. In general, improving the mechanical properties of the bioplastics depended on the amount of cellulose used to prepare these bioplastics. The film positive control is more brittle than SC₁ and SC₅ and, these are both strong and flexibles. The bioplastics that added cellulose can absorb more energy than the others, since they undergo higher strains before breaking. These results showed that the mango cotyledon starch can be considered suitable for manufacturing resistant and flexible bioplastics with similar properties to those of synthetic low-density polyethylene (LDPE), and which can be used to develop environmentally friendly materials.

3.2.2 Biodegradation

The biodegradation process of the different bioplastics can be seen in the photographic analysis shown in Figure 3 and also by measuring weight loss (Table 2), which is also considered as an indicator of degradation.

LDPE sample weight was constant and there were no observed changes in its surface, color or cracks which is an indicator for its biodegradation (Figure 3d). Exposure in soil is not enough to start the biodegradation process, giving indications that these materials remain longer in the ecosystem^[76].

It was observed that the positive control films had a greater loss of material on the 30 days, this loss of material was longer observed (Table 2). However, although it can be seen that showed the highest weight loss, the strong hydrophilicity and poor mechanical properties of the material based on starch prevent its application in the bioplastic products^[29]. As a result, it is logical to improve its properties for certain applications using cellulose as a reinforcing material.

3.2.2.1 Surface morphology by Scanning Electron Microscopy (SEM)

Only bioplastics were analyzed at different degradation times to determine a superficial analysis of the films with the aid of scanning electron microscopy since the LDPE sample did not record any changes in the percentage of biodegradation. The positive control sample showed some pores on its surface and some irregularities associated with processing operations (Figure 4a). SC₁ and SC₅ samples presented a smooth, compact, orderly surface, without the presence of pores, which indicates that starch and cellulose tend to form a homogeneous structure (Figures 4b and 4c).

Table 1. Mechanical properties of the bioplastics elaborated with starch/micro-cellulose from waste mango and polymer plastic (LDPE, negative control).

Sample	Tensile strength (MPa)	Elongation (%)	Young's modulus (MPa)
Positive control	1.465	5.667	4.655
SC ₁	4.674	24.507	11.445
SC ₅	4.961	27.950	14.350
Polymer plastic (negative control)	12.582	41.492	37.251

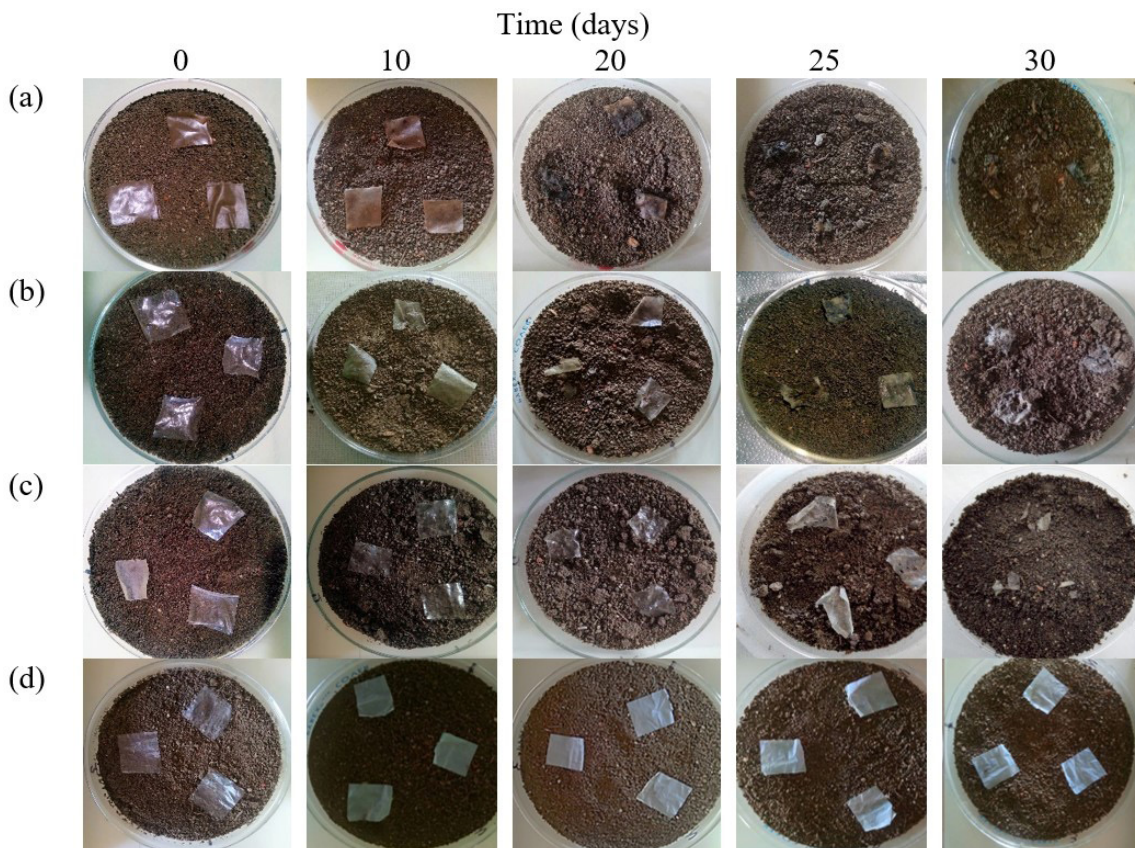


Figure 3. Photographs of samples under laboratory conditions to observe biodegradation of bioplastics samples: (a) Positive control; (b) SC₁; (c) SC₅; and (d) LDPE (negative control).

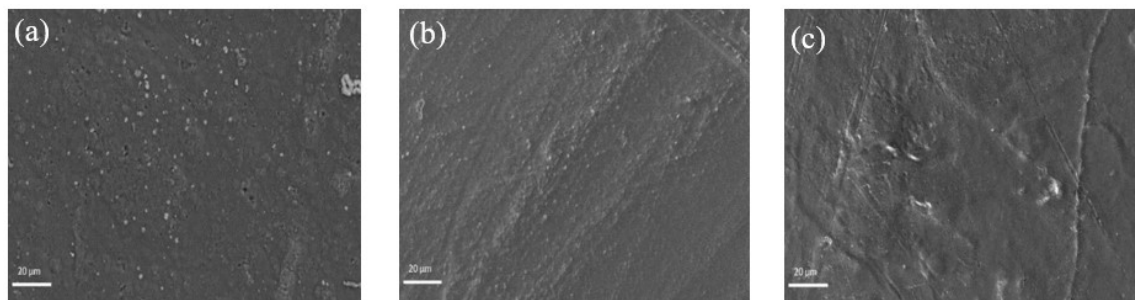


Figure 4. Surface SEM images (x 200) of bioplastics samples: (a) Positive control; (b) SC₁; and (c) SC₅ at the beginning of the biodegradation test.

Table 2. Weight (g) record of the bioplastic and LDPE samples (Negative control).

Sample	Time (days)				
	0	10	20	25	30
Positive control	0.0354	0.0230	0.0082	-	-
SC ₁	0.0362	0.0270	0.0095	0.0054	-
SC ₅	0.0325	0.0231	0.0093	0.0068	-
Polymer plastic (Negative control)	0.0044	0.0044	0.0044	0.0044	0.0044

The observation of homogeneous and compact matrices is an indication of structural integrity and consequently is expected

to have good mechanical properties^[77] in comparison with positive control sample, as presented in Table 1. Besides, the porosity and irregularities structure predicted the lower tensile strength of bioplastics^[77].

After 10 days of degradation, the bioplastics began to show the presence of fungal growth on the surface (Figure 5). In the positive control sample, mycelial growth was evident, the surface was completely covered and pores appear on the bioplastic (image a).

In Figure 5b the presence of mycelia can be observed as well as an increased presence of pores on the surface, being larger in the SC₅ sample (Figure 5c). Filamentous

microorganisms develop their mycelium within and/or on the surface of the material, causing damage to the surface of the film^[78], as revealed by the SEM images (Figure 5), also weight loss occurring in the bioplastics due to the presence of soil microorganisms is shown in Table 2 in the soil.

After 20 days of degradation, the samples showed a great impact on their structure, the positive control sample showed the presence of spores and mycelia throughout the sample and the fungal impact was also evident throughout the sample (Figure 6a).

This can be attributed to the action of starch and its high water absorption, generating a greater proliferation of microorganisms. Then, the water associated to the polymers, as observed in FTIR (Figure 2), has a significant effect which can contribute biodegradation^[79] and which is related to the predisposition to the chemical structure of the bioplastics to attack by microorganisms. Polymers like starch and cellulose are biologically synthesized and can be completely and rapidly biodegraded, either as surface erosion and bulk erosion, by heterotrophic microorganisms in a wide range of natural environment^[80]. The SC₁ and SC₅ sample, in addition, were covered by mycelia and spores, the pores are more evident at the time of biodegradation (Figure 6b and 6c) and may be due to the presence of glycerol in the polymeric matrix whose trend is to form hydrogen bonds, trap water and bond with it^[75], thus increasing soil water bioavailability which attracts microorganisms to attack the bioplastic. In the other hand, also the presence of microorganisms in the soil, responsible for carrying out microbial attack to polymer carbon backbone. As can be seen, cellulose matrices can be affected by various parameters, such as the existence and diversity of microorganisms in the soil, glycerol and microcellulose concentrations, the carbon source availability and degradation time^[75,81].

According SEM micrographs of the surface of the bioplastics was observed the biodegradation behavior and how its microstructure was mainly affected by the presence of microorganisms^[82].

3.2.2.2 Percentage of biodegradation

The percentage of biodegradation was determined by the ratio of the carbon dioxide generated from the test material and the maximum theoretical amount of carbon dioxide that can be produced from the test sample^[83]. Carbon dioxide was produced by the decomposition of the bioplastics from the action of microorganisms present in the soil as well as the enzymatic attack which degrade the glucose units that made up the structure of starch and is used as a carbon source as products of microbial metabolism for the generation of CO₂, H₂O and biomass^[83-85]. Bacteria such as: *Pseudomonas sp.*, *Streptococcus sp.*, *Staphylococcus sp.*, *Bacillus sp.*, and *Moraxella sp.*; as well as some fungi (*Aspergillus sp.* and *Penicillium sp.*) have been reported and associated with the degradation of starch-based films under soil conditions^[86].

By applying Equation 3 the percentage of biodegradation of bioplastics was obtained (Figure 7). 90% biodegradation means that 90% of the carbon atoms present in the film were converted to carbon dioxide (CO₂).

After five days, biodegradation close to 20% was observed for all starch samples; with the positive control sample having the highest value (~18%) compared to the other samples (SC₁ and SC₅). All the bioplastics tested displayed a similar biodegradation behavior; the curves in Figure 7 show the progressive increase in percentage of biodegradation as a consequence of microbial attack to polymer. This is in agreement with the photographic analysis (Figure 3) and observation of weight loss (Table 1), e.g., after 10 days the weight of the bioplastics decreased,

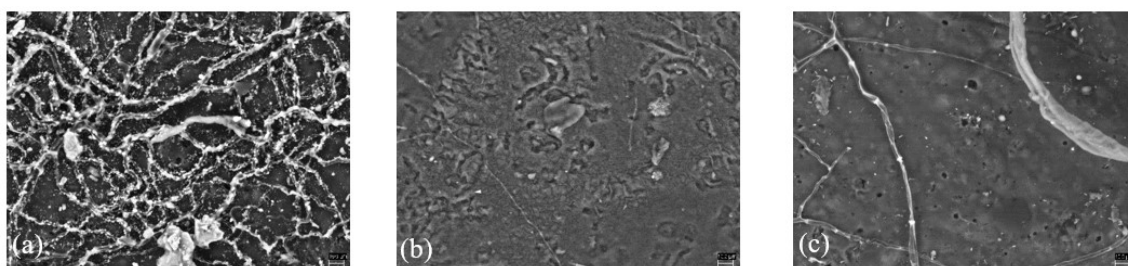


Figure 5. Surface SEM images (x 500) of bioplastics samples: (a) Control; (b) SC₁; and (c) SC₅: 10 days of biodegradation test.

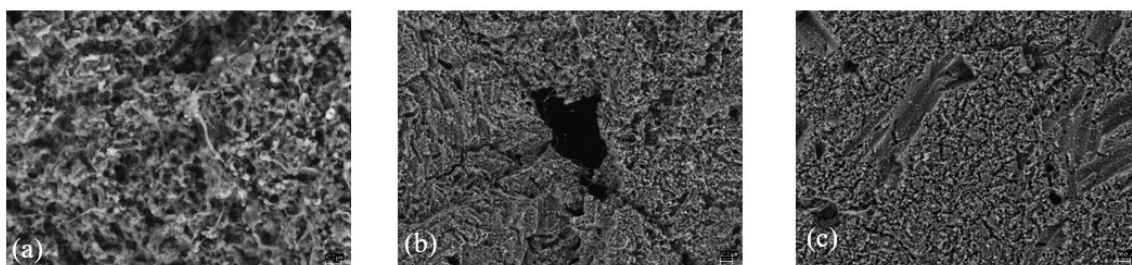


Figure 6. Surface SEM images (x 500) of bioplastics samples: (a) Positive control; (b) SC₁; and (c) SC₅: 20 days of biodegradation test.

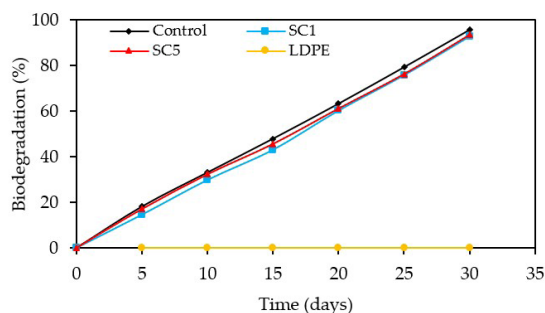


Figure 7. Biodegradation (%) of bioplastics and LDPE samples.

reaching percentage of 35, 25 and 28%, for positive control, SC₁ and SC₅, respectively. The difference in weight loss is attributed to the fact that α -1,4-glycosidic bond in starch are more easily broken down than β -1,4-glycosidic bond in cellulose^[87], regardless of what the cellulose have weak hydrogen bonds easily degraded^[88].

This behavior was constant throughout the test, reaching a maximum percentage of biodegradation of 96, 93 and 94% for positive control, SC₁ and SC₅, respectively at the end of the test (30 days). This could be attributed to the hydrophilicity of starch and glycerol, favoring the absorption and increase in water activity which promotes the growth of microorganisms^[75,81]. The value obtained after 30 days of biodegradation, possibly corresponds to the mineralization stage of the bioplastic, that is, carbon that has not been transformed into inorganic carbon, as well as to the biological activity present in the soil^[89,90]. The behavior of the other samples was consistent with the analysis presented in Figure 7 with respect to the percentages of biodegradation that can be attributed to the microorganisms. The first degradation mechanism is associated with the leaching of glycerol, which is eventually absorbed by the soil or cross the cell membrane of microorganisms, initiating the metabolism of microorganisms and increasing the biodegradation^[87,88]. It's worthy of note that the LDPE sample (negative control) did not show any change over time of the biodegradation test. These results of biodegradation of bioplastics are presented as evidence of rapid disintegration of bio-based materials and as a partial solution to the environmental problems derived from plastic waste.

4. Conclusions

Bioplastics are gaining popularity mainly due to the presentation of studies that reveal that are potentially biodegradable. In this work, the bioplastics based on cotyledon starch incorporated with micro-cellulose were developed, and the effect of cellulose on the mechanical properties and biodegradation of bioplastics were studied. The results showed that the use of micro-cellulose as a reinforcement had a positive effect on the elastic modulus and the tensile strength of the bioplastics. The biodegradation analysis showed a higher degree of disintegration of the samples in laboratory conditions as well as that this biodegradation process is very fast. The addition of cellulose increased the biodegradation time for both formulations, lasting 10 -

20 days at maximum. Once the microorganisms assimilate the biopolymer, they can degrade its structure and use it as a carbon source. Although the average degradation time was 30 days, it is necessary to carry out a greater number of variables that help us determine the capacity of micro-cellulose in other polyblends. The experiment carried out suggests that bioplastics based on biopolymers should also be tested in real conditions. Based on these findings, the waste mango can be a good choice for the low-cost obtaining biopolymers such as starch and cellulose, which are very useful for the production of plastic biodegradable. A relevant characteristic of biobased material is the biodegradation rate because this is a parameter that is necessary to try to predict the environmental fate.

5. Author's Contribution

- **Conceptualization** – Rodolfo Rendón-Villalobos.
- **Data curation** – NA.
- **Formal analysis** – Miguel Angel Lorenzo-Santiago; Roberto Olvera-Guerra; César Arnulfo Trujillo-Hernández.
- **Funding acquisition** – Rodolfo Rendón-Villalobos.
- **Investigation** – Miguel Angel Lorenzo-Santiago; Roberto Olvera-Guerra; César Arnulfo Trujillo-Hernández.
- **Methodology** – Miguel Angel Lorenzo-Santiago; Roberto Olvera-Guerra; César Arnulfo Trujillo-Hernández.
- **Project administration** – Rodolfo Rendón-Villalobos.
- **Resources** – NA.
- **Software** – NA.
- **Supervision** – Rodolfo Rendón-Villalobos.
- **Validation** – NA.
- **Visualization** – NA.
- **Writing – original draft** – Rodolfo Rendón-Villalobos; Miguel Angel Lorenzo-Santiago.
- **Writing – review & editing** – Rodolfo Rendón-Villalobos; Roberto Olvera-Guerra; César Arnulfo Trujillo-Hernández.

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