Research Article

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Composite of polylactic acid and microcellulose from kombucha membranes

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Abstract: Polylactic acid (PLA) is one of the main components of biodegradable and biocompatible composites. Bacterial cellulose from kombucha membranes is an excellent candidate to be used as a natural filler of eco-composites because it is renewable, has low cost, low density, and acceptable specific strength properties, and is biodegradable. The study aimed to prepare composites of PLA and bacterial cellulose to produce a biodegradable and compostable material. The bacterial microcellulose was obtained from kombucha membranes and blended with PLA by extrusion. The composites contained a PLA with 1%, 3%, and 5% of cellulose. We characterized the PLA, bacterial microcellulose, and composites to ascertain their size and aspect, degree of crystallinity, distribution of the cellulose into PLA, and their mechanical properties. We observed an increase in crystallinity proportional to the cellulose content for the blends and found that the 3% cellulose blend withstands the stress of up to 40 MPa and temperatures up to 120°C before distortion.

Keywords: polylactic acid, bacterial cellulose, kombucha, SCOBY, composite

1 Introduction

Environmental concerns have raised increasing interest in biodegradable polymers with mechanical properties similar to those of polyolefins. Several biodegradable polymers have recently been developed whose main uses are related to biomedical applications such as sutures, implants, drug-release systems, among others (1–3). Industrial applications of biodegradable polymers, however, are mostly limited to food packaging, probably due to their limited mechanical properties, high production cost, complex processing, and low thermal stability (4,5).

Polylactic acid (PLA) is one of the best characterized biodegradable polymers. PLA is a low, intermediate, or high molecular weight biopolymer produced either by ring-opening polymerization of lactides or condensation of lactic acid monomers produced during bacterial fermentation of agricultural products such as corn, beet sugar, and cane sugar (6). PLA has good mechanical properties, thermal plasticity, and is easy to process for paper coating, packaging, and films and fiber preparations (7). However, PLA uses are stalled by its poor crystallization rate, low crystallinity, low heat distortion temperature, and high cost (8). Biodegradable polymers are blended with other polymers to improve their mechanical properties, which, in turn, expand their applications in industry (5,9). Cellulose has proved to be a functional filler for composites and, under particular experimental conditions, has improved the mechanical properties of PLA (10,11).

Cellulose comes from different sources, such as paper pulp, cotton fibers, linen, coconut fibers, and bamboo (12). The extraction of cellulose is complicated because it is embedded within a hemicellulose–lignin matrix (13). Alternative sources of cellulose are specific microbial communities that produce highly pure cellulose, compared with vegetal sources (14). Bacterial cellulose is an industrial waste of the sweet tea fermentation known as kombucha (15). Kombucha is a symbiotic colony of
bacteria and yeast (SCOBY) composed of acetic acid bacteria (Glucanacetobacter xylinus, Acetobacter xylinoides, Bacterium gluconicum, Acetobacter aceti, and Acetobacter pasteurianus) and osmophilic yeasts (Candida sp., Kloekera sp., Schizosaccharomyces pombe, S. ludwigii, Saccharomyces cerevisiae, Torulospora sp., Zygosaccharomyces bailii, and Pichia sp.) adhered to a thick membrane of bacteria-produced cellulose (16–18).

During the biosynthesis by bacteria, cellulose chains are enzymatically polymerized from activated glucose, extruded through the bacterial cell wall, and subsequently assembled into a flexible network of cellulose microfibrils (19). Bacterial cellulose has the same chemical structure as plant-derived cellulose, but without lignin or any other contaminants. Bacterial cellulose has unique properties such as high purity, an ultrafine and highly crystalline network structure, a superior mechanical strength, pronounced permeability to gases and liquids, biodegradability, and biocompatibility (19,20). Bacterial cellulose is used in a wide range of applications from optically functional materials and composites, to novel materials for medical applications such as wound dressing and dental applications (21).

Cellulose has a vast array of industrial applications, including the paper industry, food packaging, pharmaceutical applications, and green composites formation (22). Cellulose confers thermal insulator and thermal-resistant properties when blending (23,24). Besides, crystalline microcellulose and nanocellulose possess high mechanical resistance, large surface area, chemical stability, crystallinity, transparency, biocompatibility, and electrical and magnetic susceptibility that broadens the cellulose usability (25–27).

It is common to blend the PLA with other biodegradable polymers to overcome the limitation in applicability, but it is also common to have interfacial incompatibilities. As a result, compatibilizers such as glycidyl methacrylate and polyfunctional epoxides have been added to the blends (28). Maleic anhydride (MA) is another compatibilizer useful for biodegradable polymers blends because it improves the filler–matrix interface of composites (29,30).

This study aimed to prepare a biodegradable composite of PLA and bacterial cellulose using a low-cost cellulose source. The blends are expected to be lighter, completely biodegradable and compostable, and useful for various applications. We used PLA as the matrix, and it was filled with microcellulose obtained from kombucha membranes, which are a waste product from the beverages industry. By keeping materials in use, the employment of industrial waste of good quality creates a circular economy, with positive society-wide benefits. By reducing the amounts of PLA, the carbon footprint reduces as well. The composites were characterized to ascribe their suitability for industrial applications.

2 Materials and methods

2.1 Materials

Universidad Iberoamericana, Puebla, Mexico, kindly provided kombucha. High molecular weight PLA, with a melt flow rate of 6 g/10 min (Ingeo biopolymer 2003D), was purchased from NatureWorks (Minnetonka USA). Reagent grade MA was acquired from Química Mercurio (Mexico City, México). Glacial acetic acid was purchased from JT Baker Mexico (Ecatepec, México).

2.2 Preparation of kombucha membranes

The raw cellulotic material was obtained from kombucha membranes, a by-product of the fermentation of tea broth. The tea infusion was produced from 6 g of black tea (La Pastora, México), sweetened with 114 g of table saccharose per liter of boiling water. After cooling to room temperature and removing the tea leaves, the sweetened infusion was adjusted to pH 3 with acetic acid and aseptically transferred into sterile glass bottles. The sweetened infusion was inoculated with 1 mL of fermented kombucha tea as inoculum per liter. The symbiotic colony of bacteria and yeast (SCOBY) we used contained the acetobacteria Acetobacter xylinus, Gluconacetobacter hansenii, and the yeasts Zygosaccharomyces sp., Brettanomyces/Dekkera sp., and Pichia sp. The glass bottles were covered with a cotton towel fixed to the neck of the bottle with an elastic band. The kombucha culture was fermented at room temperature (around 20–25°C) for seven weeks to produce the kombucha membranes.

2.3 Cellulose purification

The kombucha membranes were washed three times with boiling water and broken down in distilled water with a countertop blender. The water was removed by filtering,
and the cellular remnants were eliminated with 1 M NaOH for 1 h at 90°C. The cellulose was filtered using a paper filter and washed to a neutral pH with distilled water. The cellulose was further treated with 10% hypochlorite, stirred for 15 min, filtered, and washed with boiling distilled water three times. The cellulose was dried in an oven at 105°C overnight, followed by milling with a hammermill (Veyco, Mexico). The microcellulose was separated by sieving in a vibratory sifter (Montinox, Mexico) through five sequential sieves of 250, 177, 140, 125, and 37 mm.

2.4 PLA–cellulose composites

To avoid hydrolytic degradation, PLA was dried in a fan oven at 90°C for 2 h and cellulose at 60°C for 3 h before processing. Initially, a neat PLA run (used as a control) was prepared in a single screw extruder (Beutelspacher SB-19, Mexico) at 160°C and 15 Hz. The extruded material strand was cooled in a water bath and pelletized into 3–5 mm pellets. Pellets were dried at 60°C for 4 h to eliminate humidity. Then, a matrix masterbatch consisting of PLA/MA at a 99/1 weight ratio was reinforced with 1%, 3%, and 5% in weight of cellulose, by mixing in the same single screw extruder at 160°C and 15 Hz, water-cooled and pelletized. The obtained PLA/MA/cellulose pellets were extruded again to promote the shear-induced exfoliation of the cellulose in the PLA/MA matrix. The same procedure was performed on the remaining PLA/MA (0% cellulose) pellets used as control.

ASTM D638 Type V tensile dumbbell specimens were molded in a Belken SSF500-k5 injection molding machine at 190°C. Probes were injected at 190°C, with an injection pressure of 4 MPa, a mold temperature of 25°C, and a packing pressure of 3.2 MPa. After cooling for 6 s, the probes were ejected.

2.5 Characterization

2.5.1 Tensile properties

Tests were performed using a universal machine (Instron 600Dx, Massachusetts, US) at a constant displacement rate of 1 mm/min. Five replicate samples per formulation were used. The elastic modulus (MPa), ultimate tensile strength (UTS) (MPa), and elongation at break (%) were calculated. Data were analyzed with Bluehill 3.11 software (Illinois, US). We used Type V specimens, prepared according to the standard ASTMD638, whose thickness is 3.2 mm.

2.5.2 Differential scanning calorimetry (DSC)

Melting temperature ($T_m$) and glass transition temperature ($T_g$) were evaluated. Samples of 5 mg were weighed in aluminum pans and were hermetically sealed; an empty pan was used as a reference. Samples were heated from 20°C to 300°C with a 10°C/min heating rate (Pyris 1, Perkin Elmer. Norwalk, US).

2.5.3 Fourier transformed infrared spectroscopy (FTIR)

FTIR measurements were obtained in duplicates using an FTIR model with a Universal Attenuated Total Reflectance Spectrum Two (Perkin Elmer, Massachusetts, US) in the transmittance mode. Duplicates of the spectra were obtained between 4,000 and 400 cm⁻¹ at a scan rate of 30 scans and spectral resolution of 2 cm⁻¹.

2.5.4 X-ray diffraction

Samples were analyzed in an X-ray diffractometer Bruker D8 Advance (Billerica, US) equipped with Cu Kα radiation ($\lambda = 1.5406 \text{ Å}$) operating at 40 kV and 25 mA. Diffraction patterns were collected between 5° and 70° with a step size of 0.05° and a scanning speed of 3°/s. Each sample’s crystallinity was calculated using the Gaussian peak fitting method (31) and Origin software (Origin Lab Corporation, Massachusetts, US).

2.5.5 Scanning electron microscopy (SEM)

Samples of the materials were vacuum-sputtered with Au/Pd for 60 s using a Desk IV, Denton Vacuum (Moorestown, US). Samples were observed in a SEM (JSM-6390LV, JEOL, Japan) at 100×, 300×, 500×, 1,000×, 1,500×, and 3,000× magnifications using an accelerating voltage of 20 kV, a spot size of 20, and a working distance of 10 mm at 67 mA.
3 Results and discussion

The demands for eco-composites from biopolymers and natural fillers have attracted considerable interest in the industry and academic research. PLA has been used as the principal component of biocompatible blends and composites. Here, we prepared composites of PLA and cellulose using kombucha membranes as a source of bacterial cellulose. Kombucha membranes are a rich and non-expensive source of bacterial cellulose fibrils. Raw materials and composites were characterized to ascertain their morphology, degree of crystallinity, incorporation of the microcrystalline cellulose into PLA, and their mechanical properties (Figure 1).

3.1 Mechanical properties

Figure 2 shows the stress–strain curves for the neat PLA and its cellulose composites. A similar behavior between neat PLA and its composites was observed, with a linear region of approximately 0.05 mm/mm, followed by maximum stress of around 70 MPa. Then, for the neat PLA and its 1% and 3% composites, a reduction in stress was observed before fracture. For the 5% composite, fracture immediately followed the maximum stress. In Table 1, the tensile tests’ results in terms of the elastic modulus, UTS, and elongation at break are summarized.

Unexpectedly, the elastic modulus decreased with the cellulose content. Neat PLA has an elastic modulus of ~840 MPa, which reduced by 7.4%, 10.7%, and 16.5% for the 1%, 3%, and 5% composites, respectively. A similar trend was found for the UTS: ~67 MPa for the neat PLA, reduced by 2%, 7%, and 15.4% for the 1%, 3%, and 5% composites, respectively. Fracture strains for the neat PLA and 1% composite are similar (0.15 and 0.14); however, it significantly decreased for the 3% and 5% composite (0.10 and 0.10), representing almost a 30% reduction. Noteworthy, the 5% composite has higher values of standard deviation. For brittle materials, properties such as the tensile strength are highly dependent upon the surface finish of the specimen (32) which can explain the deviation of the 5% composite (the

![Image](image_url)
most brittle according to its elongation at break). This brittleness may be related to weak interactions between PLA and (not well-distributed) localized concentrations of cellulose, allowing the formation of agglomerates, discontinuities, and pores, which would affect their mechanical properties, mainly the elongation at break (33). Also, the temperature during the PLA’s double extrusion and molding process may have caused a degree of polymer degradation and an increase in the degree of crystallinity of PLA, which would also influence its mechanical properties (34–36).

Table 1 shows that at a higher cellulose content, the value of Young’s modulus decreases. For a concentration of 5%, we have an average value of the modulus of 701. In this scenario, the possible applications for these composites would be in materials with lower modulus values and for filling, but not for a reinforcing function. Examples of materials with lower values are polyvinylchloride (PVC) for semirigid extrusion–injection with a modulus of 485 MPa, thermoplastic polyurethane (TPU) with 40 MPa, and polyamide–nylon (PA11) with a modulus of 340 MPa (GRANTA EduPack software, Granta Design Limited, Cambridge, UK).

**Figure 2:** Stress–strain curve for: (a) neat PLA, (b) 1% cellulose composite, (c) 3% cellulose composite, and (d) 5% cellulose composite. Depicted are the individual curves of five replicates of each sample.
The advantage of the composites over the mentioned materials is that the composites would be biodegradable and compostable. Given the modulus of the suggested materials, there would be a margin for the increase in cellulose concentration.

3.2 Differential scanning calorimetry

Figure 3 shows the DSC thermograms of our composites. DSC was performed up to 300°C because cellulose degrades beyond that temperature (37,38). The thermograms of all the composites were similar to those of neat PLA. Both the neat PLA and the composites exhibited double melting peaks. Table 2 summarizes $T_m$, $T_g$, $\Delta C_p$, and $\Delta H$ values. MA showed no effect, as observed with the neat PLA probe vs PLA/MA pellet, whose $T_m$ and $T_g$ were similar. However, the increase in the content of cellulose caused a slight reduction in $T_g$. This reduction might be explained by the high amount of cellulose that could reduce the reticulation at the interfacial region, reducing the density of the PLA matrix (39), which may also explain the composites’ mechanical properties.

We observed that when the concentration of cellulose increased, the energy required for melting the material ($\Delta H$) also increased. This result supports the possibility for these cellulose composites to be used as insulating materials, and further studies are needed to warrant this use.

3.3 FTIR spectra

The IR spectra of the PLA–cellulose composites and control materials are depicted in Figure 4. Absorption peaks characteristics of the functional groups of cellulose are observed in Figure 4a. That is, $\text{OH}$ (3,400–3,200 cm$^{-1}$), $\text{C}=$O (1,765–1,715 cm$^{-1}$), $\text{C}=$O–C (1,270 cm$^{-1}$), and $\text{C}=$O–H (1,050 cm$^{-1}$). Absorption peaks found at 3,150–3,500 cm$^{-1}$ suggest the presence of a large number of stretching $\text{O}–\text{H}$. The absorption band found at 2,850–2,950 cm$^{-1}$ is consistent with stretching $\text{C}–\text{H}$ and $\text{C}–\text{H}_2$ of glucose units. The small peak observed at 1,645 cm$^{-1}$ indicates water absorption (37,40,41). The band between 1,420–1,430 cm$^{-1}$ denotes alkane deformation in $\text{C}–\text{H}$ and $\text{C}–\text{H}_2$, and peaks observed at 1,051–1,059, and 900 cm$^{-1}$ are associated with glycosidic units (42,43).

The neat PLA spectrum has all the individual IR band assignments described in the literature (Figure 4b), such as $\text{C}–\text{C}$ stretching at 867 cm$^{-1}$, $\text{C}–\text{O}$ stretching at 1,079, 1,126, 1,181, and 1,264 cm$^{-1}$, $\text{C}–\text{H}$ deformation at 1,377 and 1,452 cm$^{-1}$, carbonyl ester groups at 1,748 cm$^{-1}$, and $\text{C}–\text{H}$ stretching at 2,853 and 2,924 cm$^{-1}$ (44). The same peaks and bands of PLA were observed in the composites’ spectra (Figure 4c). Chemical interactions between the matrix and the filler are typically observed as changes in the IR spectrum of their composite, generally as shifting and broadening of the corresponding bands. In our composites, the amount of cellulose was not large enough to cause significant shifts in the spectra. The needle-like peak at 1,755 cm$^{-1}$ is denoted as carbonyl stretching $\text{C}=$O in the $\text{–CO–O}–$ group of PLA, and a slight shift and a widening observed at 1,756–1,760 cm$^{-1}$ in the composites indicate weak hydrogen bond formation between carbonyl groups of PLA and hydroxyl groups of cellulose (45).

3.4 X-ray diffractometry

XRD patterns were obtained to analyze the crystallinity of the components of our composites (Figure 5); the higher

![Figure 3: Comparative thermal analysis of differential scanning calorimetry (DSC).](image-url)

![Table 2: Glass transition temperature ($T_g$), melting temperature ($T_m$), specific heat ($\Delta C_p$), and enthalpy ($\Delta H$) for each sample](table-url)
the intensity of the peaks, the higher the crystallinity (46). The diffractogram of the kombucha’s purified cellulose showed the type I cellulose characteristic peaks at 2θ 14.4 and 22.7 (40). The kombucha membranes exhibited the pattern of amorphous type I cellulose (47). The control sample, neat PLA, exhibited a semicrystalline polymer pattern whose main peak was observed at 2θ 16.3.

The composites showed intense peaks of crystallinity at 16.3 (the main peak of PLA) and small peaks at 22.7 (the main cellulose peak) which increased in intensity when the cellulose content increased. Because PLA has a prolonged crystallization rate, and due to the fast cooling rate during injection molding, which did not provide enough time to crystallize, the crystallization peak of PLA is wide instead of sharp.

The crystallinity of kombucha membranes was 49.3%. However, the microcellulose prepared from kombucha membranes was 71.8% crystalline. The crystallinity of neat PLA was 58.8%. We observed an increase in crystallinity proportional to the cellulose content, 71.3%, 77.2%, and 82% for 1%, 3%, and 5% cellulose after
blending, respectively. PLA usually has a low crystallinity after practical processing, which limits its range of applications. An increase in the crystallization rate and crystallinity has been observed with blending that implies a nucleation effect (48). Here, we observed that blending with increasing concentrations of microcrystalline cellulose increased the crystallinity of the composite. The increase in crystallinity can be explained by the reduction in the \( T_g \) (49) and impacts the samples’ brittleness.

### 3.5 Morphology analysis by SEM

The morphology of kombucha membranes and the purified bacterial cellulose was analyzed by SEM. Kombucha membranes are cellulose microfibers interconnected into a tridimensional network structure (Figure 6). After purification, bacterial cellulose particles of 10–30 mm showed the aspect of microcrystalline cellulose (46).

Properties of the composites are strongly dependent on the uniform distribution of the fillers. Here, the SEM examination of the composites was performed on the fracture surfaces of the tensile specimens to investigate the distribution of cellulose in the PLA matrix. Figure 7 showed that the composites had interconnected layers with a small degree of interfacial interactions. Homogeneous distribution was observed for composites with 1% and 3% cellulose, while there was a small level of agglomeration with 5% cellulose. Together with the mechanical and thermal properties, the morphology of 1% and 3% cellulose composites suggests that these composites can potentially find applications in thermal insulation, lightweighting, electrical devices insulation, and...
energy-efficient buildings. The agglomerates in the 5% cellulose composite are indicative of weak adhesion/interaction between PLA and cellulose, generating stress concentrators, which, together with the reduction in $T_g$, explain the affectations in the mechanical properties of that formulation.

The interfacial adhesion between the PLA matrix and bacterial cellulose and PLA/cellulose composites' mechanical properties are useful if the cellulose content is up to 3%. It can withstand the stress of up to 40 MPa and temperatures between 60°C and 120°C before distortion. Also, the cellulose allows the composites to absorb energy, and the moderate heat tolerance and hydrophobicity of PLA suggest grease and liquid control.

Still, if the end product will not be subjected to high stress, the filler amount could be increased to better use waste materials.

Although we did not observe mechanical properties' improvement, the blends are still advantageous. The blends are expected to be lighter, completely biodegradable and compostable, and useful for various applications sustained within the described parameters.

Figure 7: Scanning electron microscopy of neat PLA, 1% cellulose, 3% cellulose, and 5% cellulose.
Besides, by keeping materials in use, industrial waste of good quality creates a circular economy, with positive society-wide benefits. By reducing the amounts of PLA, the carbon footprint reduces as well. The PLA composites field still needs improvement in production costs and high-temperature performance to facilitate processing.

We cannot pinpoint the best performance blend because it would require tests under specific uses and wearing conditions outside of this work scope. However, our composites have better mechanical properties than glass wool, styrofoam, and cellulose foam (according to the materials selector of GRANTA EduPack), which warrants not only biodegradable cups and utensils but also seals, wall cover, bowls, toys, containers, automotive applications, lids, and coatings, whose mechanical properties requirements are similar or lower to those of or blends. After a working life, they could be composted back to the earth.

## 4 Conclusions

The search for high-performance, lightweight, and mechanically robust materials is key to industrial and commercial applications which results in a lower carbon footprint. Here, we developed PLA–bacterial cellulose composites. The cellulose used in this work is a waste product of the kombucha industry that complies with the requirements of being renewable, low cost, low density, with acceptable specific strength properties, and of being biodegradable. More studies are necessary to improve the interaction and dispersion of cellulose into the PLA matrix to increase cellulose, which would lower costs and broaden the usability of the material.

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