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BioMask, a Polymer Blend for Treatment and Healing of Skin Prone to Acne

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The biomaterial entitled BioMask presents itself as an ideal film to be used in skin that present inflammations in their hair follicles, which can affect various body regions. Intrinsic characteristics of bacterial cellulose (BC) are important and can aid with the aesthetic treatment in the dermocosmetic area. BC's film is hypoallergenic, nontoxic, has high water retention, high purity, flexibility, biocompatibility and biodegradability. When BC is produced in a sustainable way, it can become a fundamental rich raw material for cosmeceuticals. The BioMask is enriched with bioactive compounds of high efficacy to help the healing process of the skin. Natural extracts are incorporated into dermo cosmetic products for the purpose of moisturizing, perfuming, and aggregating medicinal properties. Propolis extract is a natural waxy substance, produced by bees from the resin of several plants. Chemically, it has the presence of polyphenols and many flavonoids and innumerable biological activities. Thus, medicinal products containing propolis in the composition are used in medical treatments, and satisfactory results can be observed in the healing process and the reduction of symptoms of inflammation. The BC film was produced in Hestrin-Schramm (HS) medium and then a 2 % propolis extract was incorporated in the obtained film. The film was characterized by XRD and TGA spectroscopy. The pellicles were sterilized and a prototype of the BioMask was done. The objective of this study was to develop a BioMask that helps in the healing of inflammations caused by acne. Due to BC's high-water content, its usage as an occlusive mask can also aid in skin hydration and enable the improvement of the texture of the treated skin. BioMask is considered a dermatological and cosmetic product for immediate consumption and disposal. Being used as a possibility of treatment for acne patients, it is expected a minimization of pain, improved skin hydration and acceleration in the healing process of the skin without much aesthetic damages and improving the self-esteem and quality of life of people who have acne.

1. Introduction

Functional and advanced materials comprise a very dynamic research area in which all aspects of materials science are included, such as chemistry, physics, engineering, design, biology, and nanotechnology (Gao et al., 2019). Various types of utilitarian materials have been produced for different purposes, for example, food industry (Azeredo et al., 2019), drug delivery (Weyell et al., 2018), cosmetics (Amorim et al., 2019), treatment of oily water (Galdino et al., 2019) and others.
Such materials can be made out of bacterial cellulose (BC), which has been increasingly explored in view of its amazing chemical and physical properties, including low cost, ease, high adaptability and versatility, hydrophilicity, biocompatibility and biodegradability and non-toxicity (Campano et al., 2015).

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Active substances used in beauty care products are incorporated into excipients, which are responsible for the transportation to the skin, by permeation or penetration. Finding new natural vehicles is very important in order to make the production process less expensive and also sustainable.
Recently, BC has been deeply investigated as a support for drug release, like in applications in dental therapies with BC impregnated with the antibiotic doxycycline (Weyell et al., 2018). Or even to be used as a stabilizer for Pickering emulsions droplets in alginate beads for hydrophobic drug delivery (Yan et al., 2019). Results show its efficacy for fast release of both hydrophobic and hydrophilic drugs in a deeply satisfactory way. Propolis is a combination of resinous substances used by bees in order to defend their hive (Kedzia, 2008).
Although, raw propolis cannot be used directly in analysis or treatment. In the first place, it must be extracted so as to break down and discharge the most active ingredients. The accompanying solvents are utilized as the extractants: water, ethanol, acetone, methanol, hexane, and others (Gómez-Caravaca et al., 2006). Depending on the used solvent, different biological activity can be found (Przybyłek et al., 2019).
Propolis can have a combination of more than 300 chemical constituents such as phenolic acids, flavonoids, and caffeic acids. Also, it can contain micro and macroelements (Ca, Fe, Mn, and others) and vitamins B1, B2, B6, C and E (Pasupuleti et al., 2017) altogether, such components are known for excellent wound healing properties within anti-microbial, anti-inflammatory and regenerative activities (Altiparmak et al., 2019). In terms of antibacterial activity, the content of substances such as flavonoids and phenolic compounds is of importance (Inui et al., 2014).
Recent trends are moving toward combination of both active substances and natural vehicles in order to develop new sustainable cosmeceutical products. Regarding this idea, this work consists on the usage of a BC film, as a support for the propolis extract occlusive release, with the application as a BioMask to be used on skin prone to acne and inflammations.

2. Materials and Methods

2.1 Microorganism and Cultivation Condition

For BC production, a strain from the bacteria *Gluconacetobacter hansenii*, (ATCC 53582) obtained from the culture collection of Nucleus of Resource in Environmental Sciences, from the Catholic University of Pernambuco, Brazil, was used. The methodology for the synthesis of BC was divided into four steps: activation, pre-inoculum, inoculum and cultivation in the modified medium. The strain was maintained in the HS liquid medium described by Hestrin and Schramm (1954) which contained 2.0 % glucose (w/v), 0.5 % yeast extract (w/v), 0.5 % peptone, 0.27 % Na_2HPO_4 (w/v), and 0.15 % citric acid (v/v). BC was then produced in the modified HS medium, which has in its composition juice from the tropical fruit residue, Na_2HPO_4, and citric acid, which was adjusted to pH 6.

2.2 Activation, Pre-inoculum and Inoculum

In the activation phase, the bacteria was inoculated into HS-agar medium and incubated at 30 °C until growth, for 48 h. Afterwards, the grown cells were transferred from the activation to the pre-inoculum, which was prepared in liquid HS medium, in static conditions for 48 h, at 30 °C, then, 3 % of the pre-inoculum was transferred to the inoculum in the HS media, under the same conditions, and further experiments were done after 7 days.

2.3 Purification

After being obtained, the pellicle was washed under tap water. Then, it was purified in a 0,1 M solution of NaOH, at 90 ^{o}C for 20 minutes, in order to eliminate all retained bacterial cells, as shown. The pellicle was washed in deionized water until neutral pH was achieved.

2.4 Characterization Analysis

The films were sterilized in an autoclave at 121 ^{o}C for 15 min. Subsequently, the purified cellulose was dried at 30 °C for 30h until the reach of a constant mass as demonstrated in Figure 1. Characterization from both films, pure BC and BC and propolis extract (BC-P) was done.

Figure 1 Dried BC - P film

2.4.1 X-ray Diffractometry (XRD)

X-ray diffraction of all BC films were measured using a diffractometer (Rigaku) with Cu Kα radiation. The crystallinity index was measured by equation of Segal given below.

$$CI = \frac{(I_{002} - I_{am})}{I_{002}} .100 \quad (1)$$

In this equation, the CI expresses the relative degree of crystallinity, I_{002} is the maximum intensity (in arbitrary units) of the 002 lattice diffraction, and I_{am} is the intensity of amorphous part diffraction in the same units at 28 = 18 °.

2.4.1 Thermogravimetric Analysis (TGA)

Thermogravimetric analysis (TGA) was performed using a Mettler Toledo analyzer on samples weighing approximately 8 mg. Each sample was scanned along a temperature range from 30 ºC to 600 °C, under a nitrogen atmosphere with a heating rate of 10 °C/min and a flow rate of 20 mL/min to avoid thermoxidative degradation of the sample.

2.5 Preparation of the BioMask

The hydrated BC membranes were weighted, and half of its total mass was removed by pressure. This procedure resulted in the release of trapped water in the membrane. The membranes were immersed in a tray, and the 2 % propolis extract (distillated water was used as the solvent) was distributed at their surface. The trays were shaken for 35 minutes at 150 rpm and 30 °C. During this procedure, the membranes were turned around every 5 min in order to provide a homogenous distribution on the substance. Figure 2 shows the film before impregnation of the propolis extract (figure 2A), where it can be seen in a white coloration and in a thinner form. And figure 2B shows after total propolis extract incorporation, with a light brown coloration and a thicker pellicle.

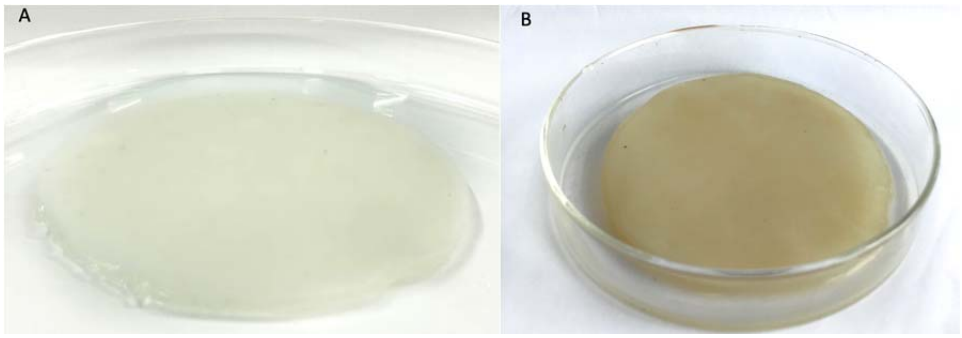

Figure 2 Pellicle before and after propolis impregnation

Afterwards, the obtained pellicles were dried at 30 °C for 20h until slightly humid (consisting on 80% of mass reduction due to water loss). The BioMask confection consisted on cutting the pellicle as the shape of other existing sheet face masks (height of 18 cm x length of 21 cm) on the market, as it can be seen on figure 3.

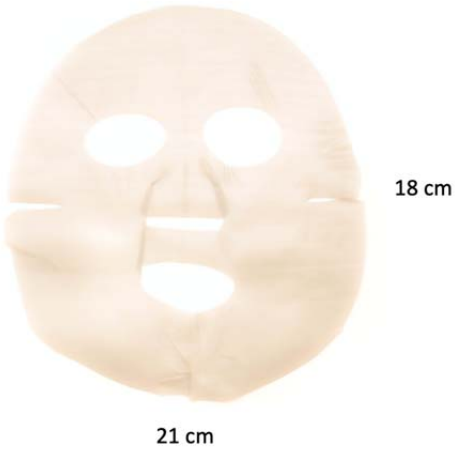

Figure 3 Prototype of the BioMask made out of BC with propolis extract

3 Results and discussion

3.1 X-ray Diffractometry (XRD)

X-ray diffraction (XRD) was used to evaluate the crystalline structure as well as the change in crystallinity of the BC with the addition of the propolis extract. An analysis obtained from BC – P diffractogram (Figure 4) shows the typical peaks of bacterial cellulose, 2θ diffraction peaks at 15.2 °, 16.8 ° and 23.6 °, which are usually attributed to the distance between the crystallographic characteristic planes, indicative of the Iα and Iβ phases of the crystalline structure (Sarma et al., 2011). As expected, both films exhibited characteristics of semi-crystalline polymers. The crystallinity index (CI) for the pure BC was of 66.7 % and for BC - P was of 46,3%. These results show that the incorporation of the propolis extract to the cellulose matrix exerted an effect on the property of the cellulose, diminishing its crystallinity.

CI has a ratio inversely proportional to the porosity of the cellulose surface (Galdino et al., 2019). Greater porosity increases the permeability of the material. So, it can be said that the decrease in the CI due to the addition of the propolis extract indicates consequently results in a higher fluid flow.

This change in crystallinity can directly affect in the mechanical properties of the membrane, reducing the tension and generating a more elastic material in comparison to pure BC. In this case, results show that BC – P has a larger amorphous region, resulting in a more flexible polymer. This malleable property enables a better application of the BioMask, because it can better adhere to the face's format.

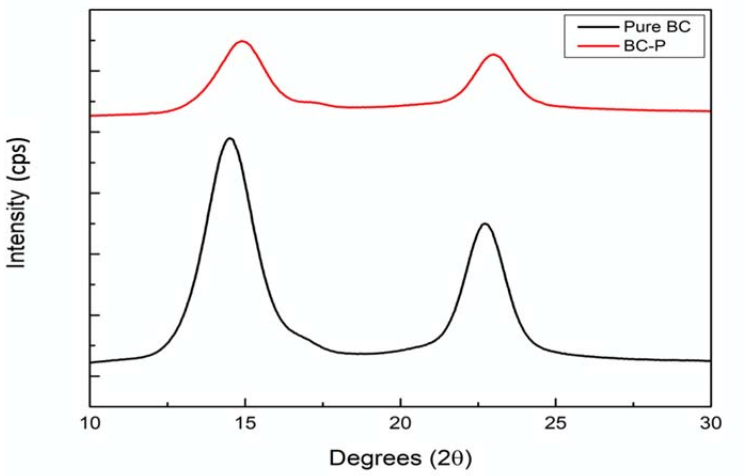

Figure 4 Diffractograms of BC obtained from pure BC and BC-P

3.2 Thermogravimetric Analysis (TGA)

The thermal stability of the BC and BC – P dry pellicles was investigated. All thermal decomposition according to its temperature stages are described in Table 1.

Table 1 Thermal decomposition of samples of pure BC and BC - P (data obtained from TGA curves)

Samples	Stage 1 (°C)			Stage 2 (°C)			Stage 3 (°C)		
	T_{onset}	T_{endset}	T_{max}	T_{onset}	T_{endset}	T_{max}	T_{onset}	T_{endset}	T_{max}
BC	25	69	55	-	-	-	294	341	324
BC - Propolis	25	129	54	149	205	191	284	344	324

The curves of the variation of mass percentage as a function of temperature are shown in Figure 5.
The TGA analysis shows that the two films showed similar degradation profiles. As the temperature increased, the mass decreased and in the BC's degradation range. The first stage is due to the evaporation of remaining water from the samples. The second stage can the attributed to the oxidative reaction of the propolis extract. As a comparison to pure BC, this additional stage can confirm the incorporation of the propolis extract on the surface of the cellulose. At higher temperatures, around 280 ºC to 350 ºC, there is another mass loss, characteristic of the thermal decomposition of the cellulose, which presents the processes of depolymerization, dehydration and decomposition of the monosaccharide units, followed by formation of carbon residues. This is the main characteristic of BC in the analysis of TGA (Barud et al., 2007).

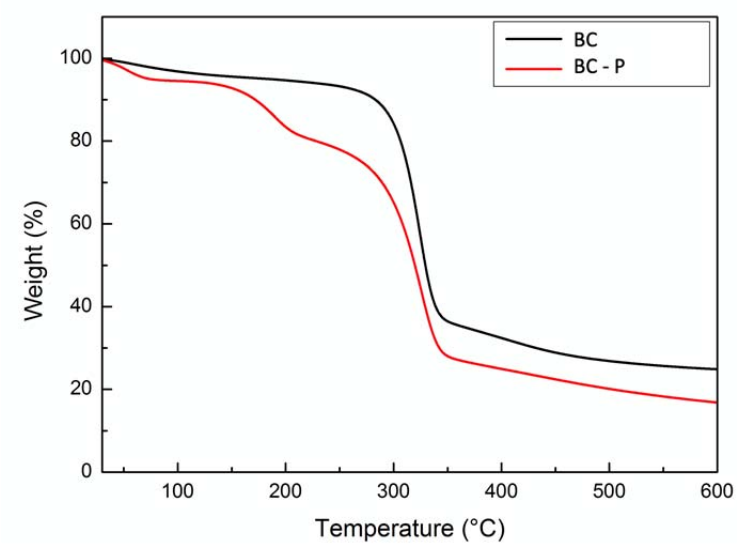

Figure 5 TG curves of both pure BC and BC - P films.

3. Conclusions

Plant and microbial natural raw materials have been used as an effective alternative in the cosmeceutical industry, because of their active natural compounds. In this study, the produced BC film was loaded with natural propolis extract to be used as a system for cosmetic skincare. The formed polymer blend presents beneficial characteristics for the usage as a BioMask in the cosmeceutical industry. The propolis extract was favourable as an autoinflammatory agent for a future application and its incorporation did not compromise the polymer's properties. This work showed satisfactory results. The incorporation of the membranes with the active did not change its nanofibrillar network structure in a critical way. It enabled better mechanical properties so that it can be used as a vehicle for releasing actives in a more efficient way. This paper describes an inexpensive and quick production process, contributing to the development of sustainable technology in the cosmetic sector. In the near future, the combination of natural biodegradable polymers, with natural active extracts can furnish new biotechnological products that meet the needs of the world market, which seeks safe and environmentally friendly options.

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