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Nanofibrillated bacterial cellulose and pectin edible films added with fruit purees

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ABSTRACT

Bacterial cellulose (BC) is a water resistant and strong material for edible films. Previous studies have been conducted on edible films containing fruit purees, but not using BC. In this study, films with or without fruit (mango or guava) purees were prepared using different ratios of nanofibrillated BC (NFBC) to pectin. The addition of fruit purees increased water vapor permeability (in about 13–18 times), reduced tensile strength (in more than 90%) and modulus (in about 99%), and increased elongation (in about 13 times), due to plasticizing effects of fruit sugars and matrix dilution by the purees. The partial or total replacement of pectin with NFBC resulted in improved physical properties, making the films stronger, stiffer, more resistant to water, and with enhanced barrier to water vapor. Fruit containing films based on pectin are suggested for sachets, whereas applications for food wrapping or coating may benefit from the use of NFBC.

1. Introduction

Edible films are self-supporting thin layers produced from film-forming edible biomacromolecules (usually polysaccharides or proteins), usually added with plasticizers such as sugars and/or polyols in order to overcome their brittleness. Casting is the method of choice to produce edible films, since the high temperatures used in other techniques are prohibitive for most biopolymers, causing degradation and lack of desirable film properties.

Edible films are usually developed to be used as food wraps, helping an external packaging in its protective function, acting then as passive protective layers without any sensory or nutritional appeal. In this context, they are supposed to be colorless and flavorless, not interfering with food sensory properties. On the other hand, sensory properties may be desirable for a variety of applications, including pouches or sachets which are melted upon cooking, wraps for sushis and sandwiches, films to separate crust and toppings of frozen pizzas, or even film snacks (Otoni et al., 2017). Since 1996 (McHugh, Huxsoll, & Krochta, 1996), several studies have described the development of edible films containing fruit purees or juices (Azeredo, Miranda, Rosa, Nascimento, & De Moura, 2012; Azeredo et al., 2009, 2016; McHugh & Senesi, 2000; Otoni et al., 2014; Rojas-Graü et al., 2006, 2007), as reviewed by Otoni et al. (2017). The polysaccharides present in fruits may act as film matrices or co-matrices, and the sugars, as plasticizers. Moreover, the market appeal of fruit-containing films is favored by the sensory and nutritional/functional properties of the fruits.

Bacterial cellulose (BC) is produced as a membrane (pellicle) by some bacteria species in a bottom-up route in which the bacteria synthesize cellulose and build up bundles of ribbon-shaped nanofibrils (Pecoraro, Manzani, Messaddeq, & Ribeiro, 2008). *Komagataeibacter xylinus* (formerly *Gluconacetobacter xylinus*) is considered as the highest producer and model organism in studies involving BC synthesis (Gullo et al., 2017). Although it has the same chemical composition as plant cellulose, BC is more pure (free from hemicellulose and lignin), which is useful to reduce purifying costs and environmental impacts (Duarte et al., 2015). Moreover, BC is free of carbonyl and carboxyl groups which are introduced to plant cellulose during purification (Nechyporchuk, Belgacem, & Bras, 2016), and has a peculiar supramolecular structure, with 3D porous networks, high degree of polymerization, high water-holding capacity, high mechanical strength, and high crystallinity (Hu, Chen, Yang, Li, & Wang, 2014; Uzyol & Saçan, 2017). Since BC is produced as a membrane, applications which require BC as a powder prior to formulation require membrane chemical and/or physical disintegration to produce nanofibrillated bacterial cellulose (NFBC) (Azeredo, Rosa, & Mattoso, 2017; Lee, Buldum, Mantalaris, & Bismarck, 2014).

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Edible films with fruit purees/juices have been usually produced with pectin (Azeredo et al., 2016; Otoni et al., 2014; Rojas-Graü et al., 2006) or alginate (Azeredo et al., 2012; Rojas-Graü et al., 2007) as biopolymer matrices. NFBC is suggested in this study as an alternative matrix, with some potential advantages to film properties due to the high tensile strength of NFBC, as well as its ability to increase water resistance of materials based on other polysaccharides (Silva, Nievola, Tischer, Mali, & Faria-Tischer, 2013; Soykeabkaew, Laosat, Ngaokla, Yodsuwan, & Tunkasiri, 2012).

In this study, films with different degrees of replacement of pectin with NFBC, added or not with fruit purees (mango or guava purees) have been produced, in order to evaluate changes in film properties derived from the presence of the fruit purees and from different degrees of replacement of pectin by NFBC as the polysaccharide matrix.

2. Materials and methods

2.1. Production of BC pellicles

BC was produced by *Komagataeibacter xylinus* ATCC 53582 in cashew apple juice, which was diluted until a sugar concentration of 20 g/L, then supplemented with yeast extract (5 g/L) and peptone (5 g/L), and its pH was adjusted to 5, in order to make it similar to the standard HS medium described by Hestrin and Schramm (1954).

The inoculum was cultivated in HS medium at 30 °C for 72 h. The supplemented cashew apple juice was cultured with 3 vol% of the inoculum and statically incubated in a BOD chamber at 30 °C for 10 days (each 500 mL of the medium on a 25 × 27 × 2 cm glass tray to produce a membrane). After fermentation, each BC pellicle was purified by washing in 700 mL of boiling distilled water for 1 h, then immersing into 700 mL of a 2% (w/v) NaOH solution at 80 °C for removal of remaining cells and culture medium. After purification, the BC pellicles were washed several times with running water until neutral pH, washed with distilled water, then autoclaved at 121 °C for 1 h and stored at 4 °C.

2.2. Production of nanofibrillated bacterial cellulose (NFBC)

The purified and neutralized pellicles were dried in an air circulation oven at 50 °C for 24 h, then ground in a Vitamix Vita-Prep® 3 blender (Vitamix Corp., Cleveland, OH, USA) at 23,000 rpm in three 1-min steps interspersed with 1-min intervals. The BC was then submitted to oxidation mediated by 2,2,6,6-tetramethyl-1-piperidinoxyl (TEMPO radical), according to the method proposed by Saito, Kimura, Nishiyama, and Isogai (2007). Ground BC (10 g) was immersed in 1 L of an aqueous solution containing 0.16 g TEMPO and 1 g sodium bromide. The oxidation was started by addition of a NaClO 11% solution (in such an amount as to achieve a NaClO concentration of 5.0 mmol/g on a dry BC basis). The pH was adjusted to 10 by adding an NaOH 0.5 M solution, and the dispersion was kept under stirring (500 rpm) for 2 h at 25 °C. After oxidation, the BC suspension was processed on Vita-Prep® 3 blender at 23,000 rpm for two 15-min steps interspersed with a 10-min interval.

2.3. Preparation of films

Two different fruit purees were used, namely: guava puree (Pomar da Polpa, Fortaleza, CE, Brazil, with 8.3 wt% total solids, 6.8 wt % total soluble solids, pH 3.5) and mango puree (Nossa Fruta, Pereiro, CE, Brazil, with 10.7 wt% total solids, 9.7 wt% total soluble solids, pH 3.5), the total and soluble solid contents and pH values being measured after filtration through 0.8-mm mesh sieves. The pectin used was from apple (Sigma-Aldrich, 70–75% methoxylation).

Three kinds of films were prepared − with guava puree, mango puree, or no fruit puree. For each kind, five films were prepared with different degrees of replacement of pectin with NFBC as the film matrix (BCPR), namely, 0, 25, 50, 75, and 100. Each film was prepared using 475 mL of fruit puree (or distilled water, for no-fruit-puree films), 0.57 g sorbitol (plasticizer), and 2.85 g matrix on a dry basis (NFBC and/or pectin), always considering that the NFBC suspension had a 1% (w/v) concentration. The film components were blended on Vita-Prep® 3 at 23,000 rpm for 10 min. The dispersions were vacuum degassed with a vacuum pump (V-700, Büchi, Flawil, Switzerland) at 30 mbar for 45 min, then cast onto 34 × 24 cm borosilicate glass containers in amounts estimated to produce films with final (dried) thicknesses of about 100 μm. The cast dispersions were then dried in an air-circulating oven (Quimis® Q-31) for 24 h at 50 °C.

Dried film samples were cut and detached from the surface. Before film characterization, the free-standing samples were conditioned for at least 40 h at 23 °C and 50% RH before characterization.

2.4. Film characterization

The film colors were determined in the CIELab color space by using a colorimeter (Konica Minolta CR-400, Konica Minolta, Osaka, Japan). Measurements were taken at five distinct points of each film sample.

Tensile properties of 125 mm × 12.5 mm film strips (with ten replicates) were measured according to D882-12 (ASTM, 2016), using an Emic DL-3000 Universal Testing Machine with a load cell of 10 N, initial grip separation of 100 mm, and crosshead speed of 12.5 mm/min.

The insoluble matter determination (used to assess water resistance) was carried out in quadruplicate, based on the method proposed by Pena-Serna and Lopes-Filho (2013). Previously dried (105 °C, 24 h) and weighed film disks (2 cm in diameter) were immersed into 50 mL distilled water for 24 h under stirring (76 rpm) at 25 °C in an orbital shaker (SL 222, Solab, Piracicaba, Brazil). The dry weight of the remaining film pieces was obtained after filtration on previously dried and weighed filter paper, and was used to calculate the insoluble matter as a percentage of the initial dry weight (g/100 g).

The water vapor permeability (WVP) determination, with eight replicates, was based on the method E96/E96M-16 (ASTM, 2016) at 25 °C, using silica gel as desiccant (0% RH) in an Arsec DCV-040 vertical desiccator (outside the permeation cells) and water (100% RH) inside the permeation cells. Eight measurements were taken within 24 h.

Fourier-transform infrared spectroscopy (FTIR) spectra were recorded in a Schimatzu Prestige-21 spectrophotometer, using KBr pellets, in the wavenumber range from 4000 to 400 cm^{-1}, with a resolution of 4 cm^{-1} over 64 scans.

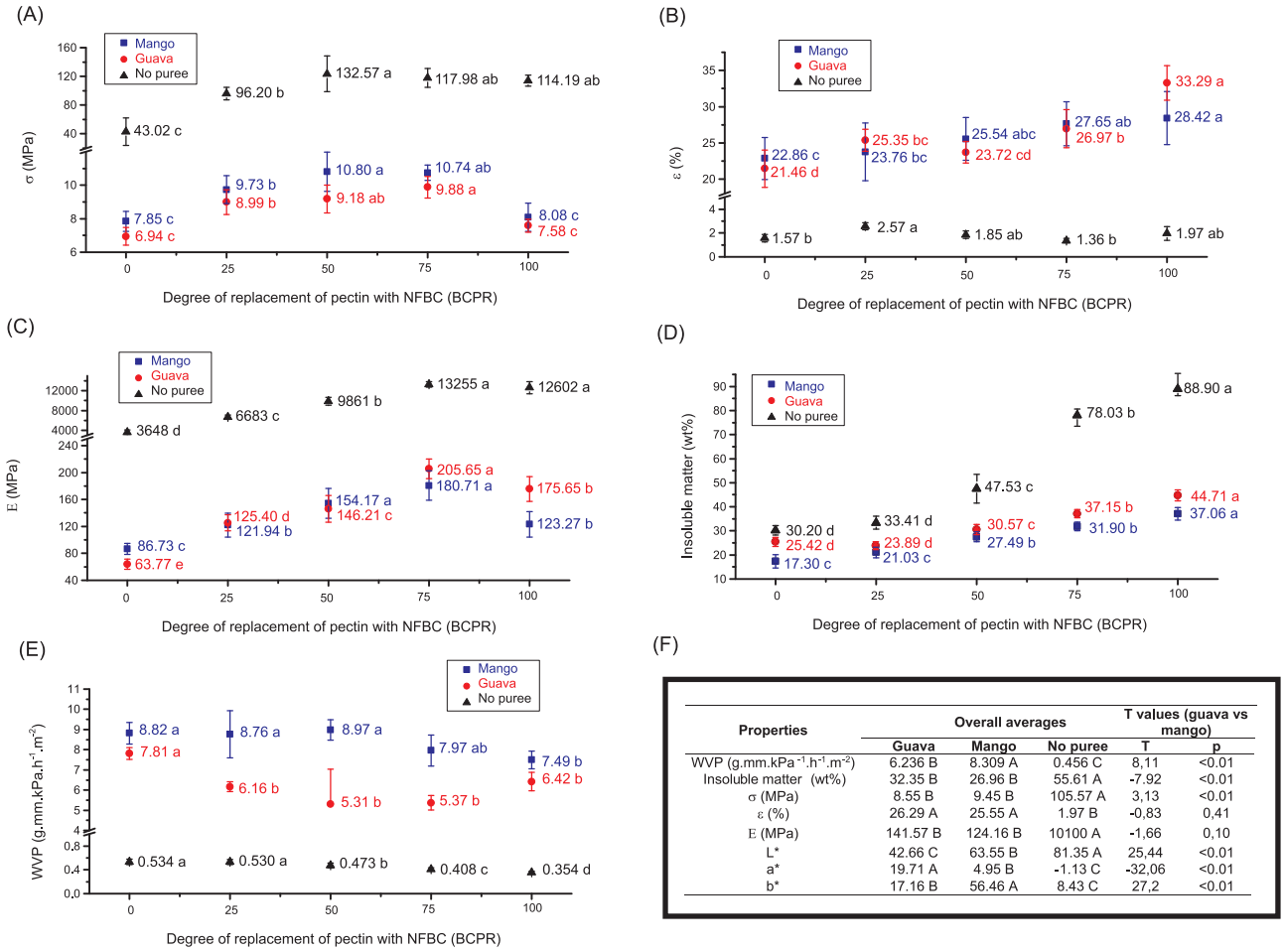
The scanning electronic microscopy (SEM) micrographs of the films were taken using a QUANTA FEG 450 microscope. The samples were mounted on the aluminum stub using carbon-coated double sided adhesive tape, gold coated, and examined using an accelerating voltage of 5 kV and a magnification of 3000 times.

3. Results and discussion

3.1. Tensile properties of films

When compared to the films without fruit purees in overall terms (Fig. 1F), the films containing guava or mango purees presented remarkably lower tensile strength and modulus, and higher elongation, which results from both the mere matrix dilution by the fruit components and the plasticizing effect of low MW sugars and acids from fruits, as previously observed (Azeredo et al., 2016; Otoni et al., 2014). When compared to films with guava puree (by paired *t* test), those with mango puree were stronger, which may be ascribed to the fibrous nature of mango puree, the fibers reinforcing the films.

When considering films with different matrix compositions (Fig. 1A–C), films with no purees exhibited maximum tensile strength at BCPR of at least 50, and maximum modulus, at BCPR of at least 75, while elongation was not much affected by the matrix composition. When compared to the pectin-only film, the NFBC-only film was about



Properties	Overall averages			T values (guava vs mango)	
	Guava	Mango	No puree	T	p
WVP (g.mm.kPa^{-1}.h^{-1}.m^{-2})	6.236 B	8.309 A	0.456 C	8,11	<0.01
Insoluble matter (wt%)	32.35 B	26.96 B	55.61 A	-7.92	<0.01
σ (MPa)	8.55 B	9.45 B	105.57 A	3,13	<0.01
ε (%)	26.29 A	25.55 A	1.97 B	-0,83	0,41
E (MPa)	141.57 B	124.16 B	10100 A	-1,66	0,10
L*	42.66 C	63.55 B	81.35 A	25,44	<0.01
a*	19.71 A	4.95 B	-1.13 C	-32,06	<0.01
b*	17.16 B	56.46 A	8.43 C	27,2	<0.01

Fig. 1. Tensile properties (A, B, and C), insoluble matter (D), and water vapor permeability (E) of films with variable BCPR values, and overall averages (F). For graphs (A-E), means not followed by the same letter within a sequence are significantly different ($p < 0.05$). For overall averages (F), means not followed by the same letter within a row are significantly different ($p < 0.05$).

2.6 times stronger and 3.4 times stiffer. Such findings indicate that the overall tensile properties were favored by the prevalence of NFBC over pectin. Gu and Catchmark (2014) found that, when cultured with pectin or xyloglucan, BC composites exhibited lower modulus than those of BC-only films, which the authors ascribed to reduced entanglement of cellulose nanofibrils by the additional polymer.

For fruit puree-containing films, the results were slightly different. Films with BCPR between 50 and 75 presented highest strength and modulus, being 30–40% stronger and up to 2–3 times stiffer than the ones without NFBC. On the other hand, films whose only matrix was NFBC (BCPR 100) presented similar strength to films whose only matrix was pectin (BCPR 0), and their modulus, although still significantly higher than those of BCPR 0 films, were decreased when compared to those at BCPR 75, whereas their elongation was further increased. Although NFBC by itself has been proven to be stronger and stiffer than pectin (from the results in no-puree films), the decreased strength and modulus of fruit-containing BCPR 100 films as compared to BCPR 75 may be ascribed to weaker interactions between the fruit purees and cellulose when compared to those interactions in presence of some added pectin, which may be due to the presence of considerable amounts of pectin in fruit purees themselves, and consequent high chemical affinity of those purees with pectin.

3.2. Water resistance (insoluble matter) of films

When compared to no-puree films, fruit-containing films presented lower overall water resistance (Fig. 1D, F), since the composition of both purees contains mostly water soluble compounds. Mango films exhibited lower insoluble matter than guava films, which is attributed to the higher (predominantly water soluble) solid content of the mango puree (See item 2.3), contributing more for the total solid content of the films, thus affecting more the insoluble matter content of the matrix.

The insoluble matter of no-puree films was deeply affected by the matrix composition (Fig. 1D), the NFBC-only film presenting a threefold increase in insoluble matter when compared to the pectin-only film. Lindman, Karlström, and Stigsson (2010) have discussed cellulose as an amphiphilic molecule, and ascribed its low solubility in water not merely to strong inter- and intramolecular hydrogen bonds, but mainly due to hydrophobic interactions.

For fruit-containing films, the higher the NFBC content, the higher was the water resistance also, although the differences were less remarkable, because of the high content of water-soluble fruit components diluting the matrix. Still, the insoluble matter contents of fruit-containing films at BCPR 100 were about twofold those of the corresponding films at BCPR 0.

The increased water resistance provided by replacement of pectin with NFBC may represent a great advantage for several applications, since the films are less susceptible to disintegration upon exposure to a moist surface. Actually, several studies have proposed methods for enhancing the water resistance of polysaccharide films, usually by crosslinking (Azeredo et al., 2015; Zhang, Xu, Gao, Fu, & Zheng, 2017) or by addition of hydrophobic compounds (Manrich et al., 2017; Santos et al., 2014). The natural water resistance of cellulose is especially advantageous, since it does not require any modifications to decrease

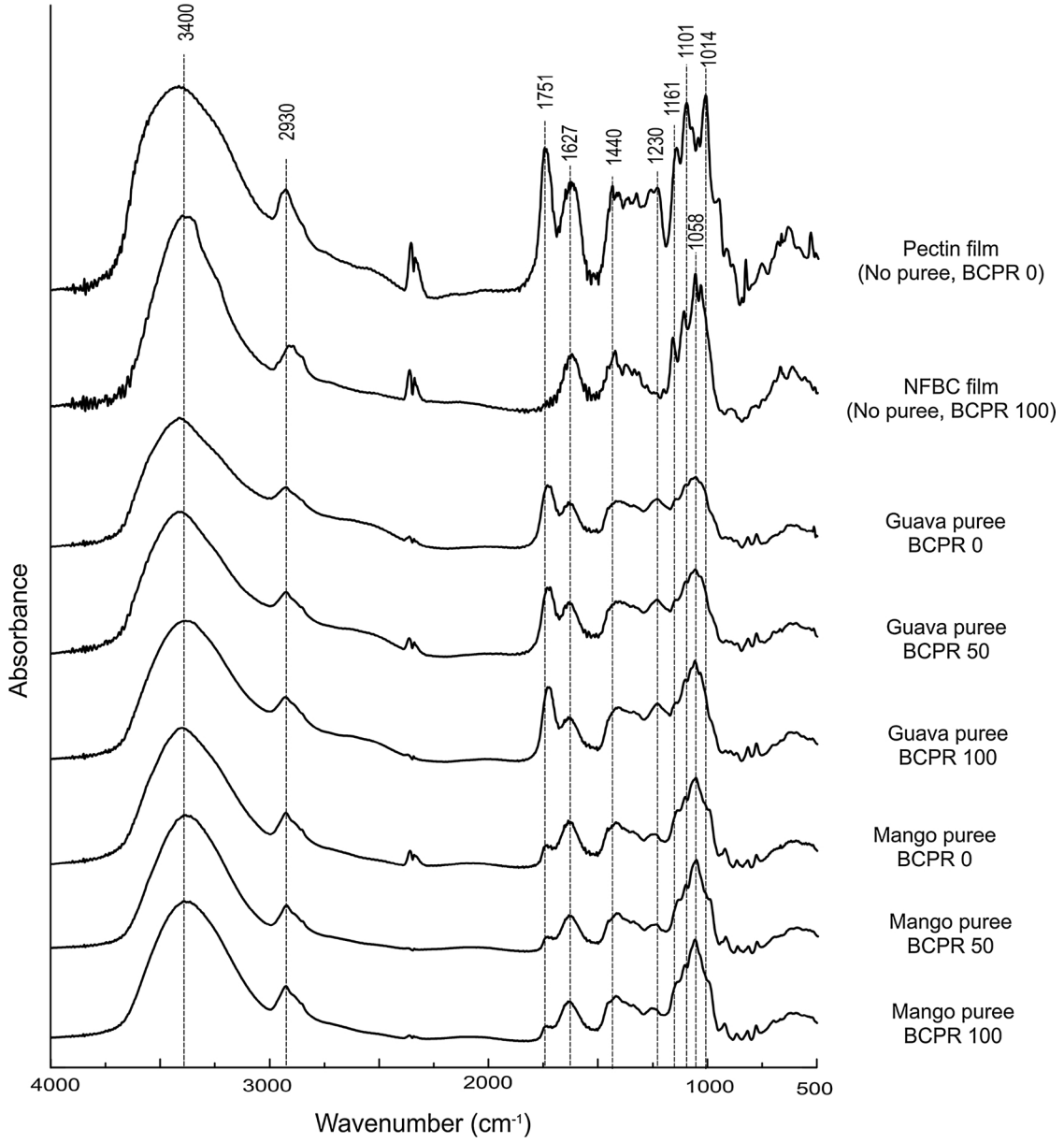


Fig. 2. FTIR spectra of films.

its water solubility.

On the other hand, some applications may require the films to dissolve in water, such as when films are used to produce sachets to release a content when exposed to water, or when flavored films are to be fully dissolved to become part of a beverage. For such cases, pectin or other water soluble polysaccharides are more suitable as matrices.

3.3. Water vapor permeability (WVP) of films

Fruit-containing films presented higher WVP than no-puree films (Fig. 1E, F), which is ascribed to the already mentioned higher solubility of fruit-containing films, since permeability is a function of both solubility and diffusivity. Films with guava puree were less permeable than those with mango puree, probably because guava puree, having a lower solid content, contributed less for the solid matter of the films, thus affecting less the barrier properties of the matrix.

The WVP of no-puree films decreased with increasing NFBC contents, which is probably due to the high crystallinity of NFBC. When compared to films without NFBC (BCPR 0), mango films exhibited significantly lower WVP values only at high NFBC contents (BCPR of 75 or more), whereas guava films presented lower WVP values at any NFBC content (from 25).

The enhanced water vapor barrier conferred by increasing NFBC contents is advantageous for most food packaging applications, since water transfer through a packaging film may result in changes in food texture and even promote microbial growth.

3.4. FTIR spectra of films

The FTIR spectra (Fig. 2) present some common bands to all films, such as a broad absorption band around 3400 ccm^{-1} (O—H stretching of hydroxyl groups), the band around 2930 cm^{-1} (C—H stretching), and the one at about 1160 cm^{-1}, ascribed to asymmetric C—O—C stretching (Colom, Carrillo, Nogués, & Garriga, 2003). The band around 1620 cm^{-1} in the NFBC film, ascribed to carboxylate groups (Coseri et al., 2015), reveals the oxidation of cellulose by TEMPO. The pectin naturally present in guava puree presented higher degree of methoxylation than the pectin from mango puree, as evidenced by the

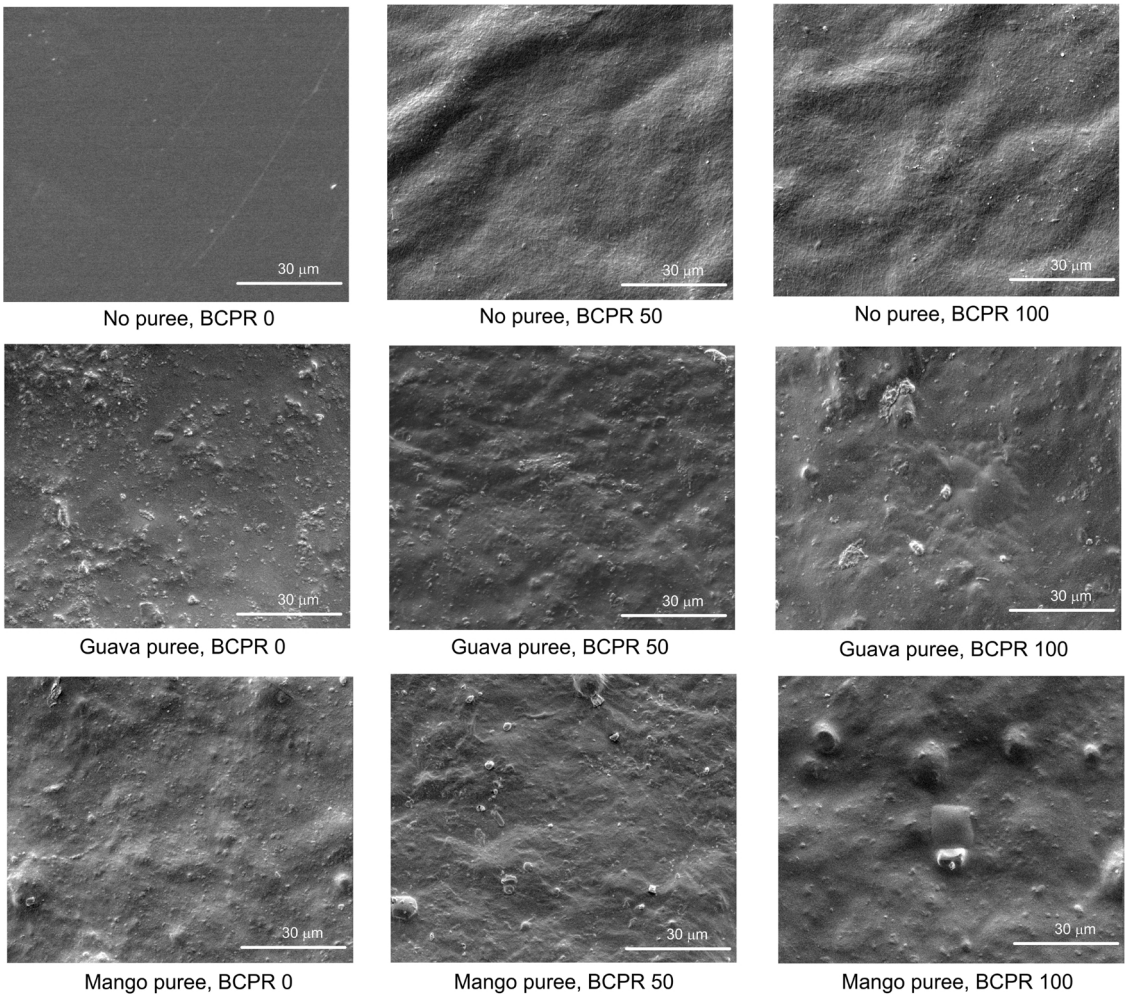


Fig. 3. SEM micrographs of the films.

difference in peak intensity ratios of free carboxyl (around 1630 cm^{-1}) to ester carbonyl (around 1730 cm^{-1}) of mango and guava films. The band around 1440 cm^{-1} are assigned to asymmetric stretching of CH_3 of pectins (Fellah, Anjukandi, Waterland, & Williams, 2009), whereas a close band at around 1430 cm^{-1} is assigned to CH_2 scissoring motion of cellulose I (Deng, Tan, Fang, Zhang, & Wang, 2009). Some bands are present in all films containing pectin (commercial pectin and/or fruit purees), such as the one at 1230 cm^{-1}, ascribed to C—O stretching of carboxylic acids (Stuart, 2004), and the ones at 1100 cm^{-1} and 1014 cm^{-1}, assigned to C—O and C—H stretching (Oliveira et al., 2016), whereas the band at 1058 cm^{-1}, assigned to the bending of C—O—C pyranose rings in cellulose (Mandal & Chakrabarty, 2011) is more prominent in films containing NFBC. The spectra do not present evidences of significant interactions between film components.

3.5. SEM micrographs of film surfaces

The SEM micrographs (Fig. 3) show noticeable differences in film surfaces. The pectin-only film exhibited a smooth surface, while the presence of NFBC provided the films with a mesh-like morphology and an undulated surface. The most noticeable change, however, was conferred by the presence fruit purees, which produced heterogeneous and rough surfaces, due to the complex nature of the fruit purees.

4. Conclusions

Films containing mango or guava puree were demonstrated to exhibit remarkable differences in tensile properties, water vapor barrier and water resistance, when compared to the corresponding films without any fruit purees. Moreover, the partial or total replacement of pectin with nanofibrillated bacterial cellulose (NFBC) resulted in improved physical properties of edible films (both of those with and without fruit purees), making them stronger, stiffer, more resistant to water, and with enhanced barrier to water vapor. Films with higher NFBC contents are proposed for applications which require some water resistance and better tensile properties, such as for food wrapping or coating, while films with only pectin as matrix is more suitable for applications which require water dissolution, such as sachets.

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