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Valorization of mangoes with internal breakdown through the production of edible films by continuous solution casting

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ABSTRACT

Mangoes are usually wasted by consumers due to a physiological disorder known as Internal breakdown (IB). In this study, Palmer mangoes affected by IB were valorized by turning into edible films by continuous solution casting. The films were produced using mango pulps with different IB levels, being 0 (no IB), 2 (1/3–2/3 of IB-affected pulp) and 3 (more than 2/3 of IB-affected pulp), and pectin as a matrix former. The influence of IB level on the physicochemical composition of mango pulps was assessed by pH, soluble solid content, and fiber content analyses. Continuous casting allowed for high productivity of edible films with natural mango color attributes. The films obtained from mango pulps with the highest IB level displayed lowest thickness and water vapor permeability, and largest elongation and opacity, in addition to exhibiting a short composting time (10 days). The physical properties of mango edible films are influenced by the pulp physicochemical composition which changes with IB progression.

1. Introduction

Mango (*Mangifera indica* L.) is one of the most important tropical fruits worldwide, mainly due to its large production (FAO, 2019), consumption (pleasant aroma and taste) and nutritional value (Ntsoane, Zude-Sasse, Mahajan, & Sivakumar, 2019). During postharvest, mangoes usually develop a physiological disorder known as internal breakdown (IB). This disorder refers to one or more physiological processes induced by environmental factors that accelerate the mango pulp ripening in a premature and uneven way, leading to a collapse and appearance of dark watery stains in the pulp (Raymond, Schaffer, Brecht, & Crane, 1998; Wainwright & Burbage, 1989). IB reduces the sensory quality of mangoes, causing a significant economic loss, not only for the mango supply chain, but also for consumers due to low availability, high price, and inferior quality of mangoes in the market (Varian & Jordan, 1988).

Collapsed mangoes are often discarded because IB still remains unknown for most consumers. The disposal of aesthetically imperfect mangoes represents a waste of fruits that still contain high nutritional

values and could be used for other purposes, thereby reducing food waste (Okawa, 2015; Parfitt, Barthel, & Macnaughton, 2010). As an alternative, collapsed mangoes could serve as raw materials for edible film production (Viana, Sá, Barros, Borges, & Azeredo, 2018).

Edible films have attracted significant interest within the perspective of replacing non-biodegradable polymers with compostable materials (Kaya et al., 2018). Recent researches have demonstrated the potential of edible films containing purees or fruit juices (Azeredo et al., 2016; Tran, Roach, Nguyen, Pristijono, & Vuong, 2020; Viana et al., 2018). Fruit components play different roles in the films, for instance, polysaccharides and low molecular weight sugars can act as matrix formers and plasticizers, respectively, while the bioactive compounds of fruits provide the films with unique sensory properties (Viana et al., 2018). In this sense, valorizing collapsed mangoes by turning into edible films is aligned with sustainability concepts, but it is also a suitable approach to develop innovative edible packaging (Song et al., 2017).

This study aimed to develop edible films from mangoes affected by internal breakdown (IB) by solution casting. Fruits affected by this physiological disorder have a gelatinous aspect, are sweeter, and often

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have a lower fiber content (Brecht, 2019; Raymond et al., 1998). In a general view, IB-affected mango pulps could form a more flexible and thinner edible film due to their higher sugar and lower fiber contents, respectively, in addition to providing mango sensory attributes, such as color and flavor, to make the film more appealing for edible packaging applications. Thus, we hypothesized that films produced from mangoes affected by IB could also meet the requirements of a food packaging material concerning mechanical, barrier and optical properties.

To obtain films, pectin was used as a matrix-forming aid, as it can be easily solubilized without heating to be further mixed with IB-affected mango pulps. More importantly, pectin was expected to form cohesive structures with the mango pulp by hydrogen bonding in the casting/evaporation process due to its hydrophilic nature (Janjarasskul & Krochta, 2010). Pectin films display good mechanical properties, and high barrier against oils, aromas, and oxygen. However, these films have low resistance to moisture and are brittle (Giancone et al., 2011). In this case, the IB-affected mango pulp could also be a good alternative to modify these gaps in the functions of pectin (Baldwin, Hagenmaier, & Bai, 2012; Sothornvit & Pitak, 2007), as the plasticizing effect of sugars present in the pulp could modify the interactions in the film matrix, thus increasing the free volume and mobility of polymer chains (Nogueira, Soares, Cavasini, Fakhouri, & Oliveira, 2019; Singh, Chauhan, Agrawal, & Mendiratta, 2018). The addition of pulp reinforces the film, making them more flexible for packaging purposes (Azeredo et al., 2016).

The study of the feasible large-scale production of films from mangoes affected by physiological disorders is also of great importance. To put this industrial perspective herein, continuous solution casting was applied to scale up the production of the IB-affected mango films. This processing method has been recently proved to be effective in obtaining edible films from wasted fruits in a soft and highly productive fashion (Munhoz et al., 2018). Hence, the mechanical, optical, and water vapor barrier properties of the continuously cast IB-affected mango films were particularly examined with respect to the IB level. No report has been published so far on these correlations. In this sense, the physico-chemical parameters of mango pulps with different IB levels were assessed and correlated with the physical properties of the ensuing edible films. The susceptibility of collapsed mango films to biodegradation was also qualitatively scrutinized by indoor composting tests.

2. Materials and methods

2.1. Materials

Mangoes (*Mangifera indica* L.) 'Palmer' ('Type 12' classification - 12 fruits in a 6.0 kg box) (CEAGESP, 2017) were purchased from CEAGESP-SP, Brazil. The fruits were selected, sanitized, and stored at 15 °C in a cold chamber until they reached the proper ripeness degree for consumption in terms of skin color, softening, and flavor (Tharanathan, Yashoda, & Prabha, 2006). The mangoes were cut and classified according to the absence or presence of IB. The fruits were further classified by the IB damage level, being: without IB (WIB), level 2 (IB2, 1/3–2/3 of IB-affected pulp) and level 3 (IB3, more than 2/3 of IB-affected pulp), as shown in Fig. 1 (Brecht, 2019). Citrus pectin with an esterification degree (DM) of 74% and an average molecular weight (M_w) of 1.3×10^5 g mol⁻¹ was purchased from CPKelco (Niplan Engenharia, Limeira, Brazil).

2.2. Pulp processing and characterization

Three mango films were obtained from pulps classified according to the IB level (WIB, IB2 and IB3). The fruits were homogenized and processed in a centrifuge to isolate the pulp from the juice into two different portions. The pulps were weighed and evaluated for their soluble solid contents (°Brix) using an Atago RX-5000cx (Honcho, Itabashi-ku, Japan) bench refractometer, pH using a QX 1500 QUALSTRON (Hexis Científica, Jundiaí, Brazil) pH meter, and total dietary fiber content as reported

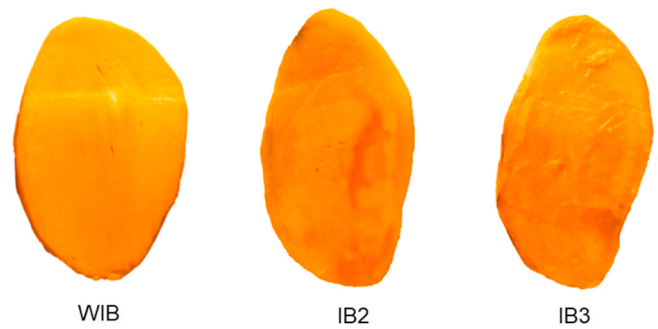


Fig. 1. Visual symptoms of jeely seed in 'Palmer' mango pulps. Fruits without (WIB), level 2 (IB2), and level 3 (IB3) of internal breakdown (IB). Photo: Fernanda, C. A. Oldoni (2019).

by Prosky et al. (1985) (AOAC, 985.29 enzymatic-gravimetric). The pulps were subsequently kept in a freezer (−20 °C) to preserve their sensory attributes.

2.3. Continuous solution casting

The edible films were produced from filmogenic solutions composed of mango pulps added with 5 wt% pectin on a pulp mass basis. The filmogenic solutions were added to a polyacetal vessel ($V_{total} = 2.6$ dm³) and vigorously homogenized at 25 °C and 7168g for 10 min using an ultraturrax. The solutions were degassed by applying vacuum at −400 mmHg alongside mechanical stirring, and were immediately turned into monolayer films on a KTF-B laboratory coating machine (Werner Mathis AG, Zurich, Switzerland). Briefly, the solution was deposited on the coating device equipped with a doctor blade type B, in which a wet solution layer with thickness of 1.5 mm was formed using a comparative dial pair (± 0.001 mm). The wet layer was carried at a speed of 12 cm min⁻¹ through an IR pre-dryer with emission power of 40–55%, then through an air circulation oven (total length of 80 cm) at 100 °C. The films were collected at the oven outlet and stored for further characterizations. Fig. 2 shows the steps of manual feeding, wet solution layer transport, pre-drying, and formation of dried mango film using the continuous casting machine. A pure pectin film (no mango pulp

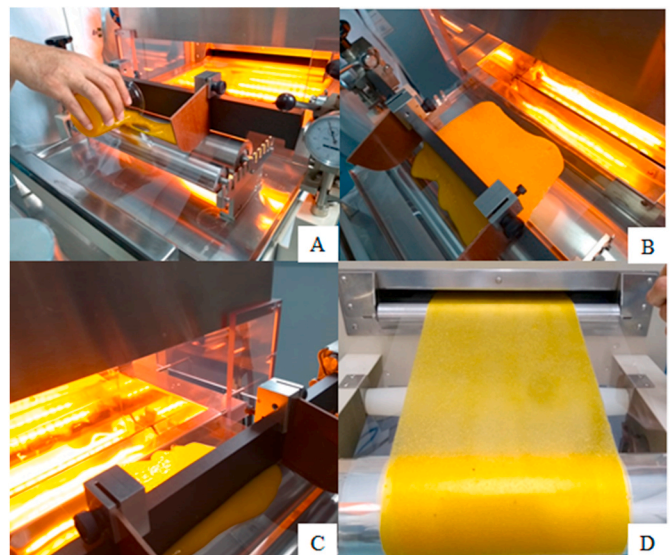


Fig. 2. Elaboration of edible mango pulp films by continuous solution casting: manual feeding of filmogenic solution (A); formation of wet solution layer at the coating device with thickness control (1.5 mm) (B); Pre-drying using IR radiation (C); Monolayer film obtaining after the last drying step (D). Photo: Fernanda, C. A. Oldoni (2019).

addition) was also prepared and used as a control.

2.4. Characterizations

2.4.1. Scanning electron microscopy (SEM)

Film morphology was assessed using a scanning electron microscope JEOL-JSM 6510 (Jeol, Tokyo, Japan). The films were conditioned in a desiccator with silica gel, cut into small pieces, mounted on a stub with an adhesive carbon tape, and finally coated with a thin gold layer. The micrographs were taken with an accelerating voltage of 15 kV and magnification of 1000 \times .

2.4.2. X-ray diffraction (XRD)

X-ray diffraction analyses were carried out on a XRD-6000 diffractometer (Shimadzu, Kyoto, Japan) with 2θ range between 5 and 40 $^\circ$ and scanning speed of 1 $^\circ$ min $^{-1}$. The crystallinity index was determined from the ratio between the areas under the peaks and amorphous halo in the diffractograms after Gaussian deconvolution using the Origin software, version 6.0 (Origin Lab, Northampton, MA, USA).

2.4.3. Fourier transform infrared spectroscopy (FTIR)

The FTIR spectra were determined with a Cary 630 FTIR (Agilent, Santa Clara, USA) spectrometer using the attenuated total reflectance (ATR) mode. The spectra were collected in the wavenumber range from 4000 to 400 cm $^{-1}$ with resolution of 4 cm $^{-1}$.

2.4.4. Thickness and moisture content

Film thickness was determined with a portable digital micrometer (Mitutoyo Co., Kawasaki-Shi, Japan) to the nearest 0.001 mm. The measurements were carried out in at least three points of each film. Square-shaped (2 cm 2) film samples were weighed before and after drying at 105 $^\circ$ C for 24 h, and the moisture content was calculated.

2.4.5. Water vapor permeability (WVP)

The WVP of films was determined in triplicate ($n = 3$) using the classical gravimetric method (ASTM E96-E96M, 2016). The films were sealed onto permeation cups (35 mm in diameter) containing 6 mL of distilled water (RH = 100%). The cups were then placed in an air circulating oven (Solab SL-102, Piracicaba, Brazil) at 40 $^\circ$ C containing activated silica gel (RH = 0%). The weight of each cup was measured with an analytical scale at least 10 times over 32 h. WVP (g mm/h cm 2 Pa) was calculated according to Eq. (1).

$$WVP = WVT \cdot \frac{x}{A \cdot \Delta p} \quad (1)$$

where WVT is the water vapor transmission rate (g s $^{-1}$ m $^{-2}$), determined as the slope of the mass loss (g) vs. time (h) curves, A is the film permeation area (m 2), x is the film thickness (mm), and Δp is the water vapor pressure gradient between the inside and outside of the permeation cup.

2.4.6. Tensile tests

Uniaxial tensile tests were performed with 10 replicates ($n = 10$) for each film sample using a Texture Analyzer TA. XT Plus (Stable Micro System, Godalming, UK) equipped with a 50 N load cell and tensile clamps initially separated by 20 mm. The tests were performed on strip specimens (50 mm \times 10 mm) using a crosshead speed of 80 mm min $^{-1}$ (ASTM D882-97). The stress-strain curves were plotted for each specimen for determining the maximum stress (σ_{max} , MPa) and elongation at break (ϵ_{max} , %) of films.

2.5. Optical properties

The color of films was determined on a CR 400 colorimeter (Konica Minolta, Osaka, Japan) using the CIELab system (L^* - brightness, a^* -

red-green, and b^* - yellow-blue) (Abbott, 1999; Pathare, Opara, & Al-Said, 2013). The hue angle (h°) which represents the qualitative attribute of color was calculated using Eq. (2) (McGuire, 1992; Shewfelt, Thai, & Davis, 1988). The chroma index (C^*) (Pathare et al., 2013; Shewfelt et al., 1988) which is the quantitative color attribute, and the total color difference (ΔE^*) (Pathare et al., 2013) were calculated using Eqs. (3) and (4), respectively.

$$h^\circ = \tan^{-1}(b^*/a^*) \quad (2)$$

$$C^* = (a^{*2} + b^{*2})^{1/2} \quad (3)$$

$$(\Delta E = \sqrt{(L^* - L)^2 + (a^* - a)^2 + (b^* - b)^2}) \quad (4)$$

The optical properties (total luminous transmittance, haze, and clarity) of films were determined according to ASTM D1003 (2007) using rectangular specimens and a BYK-Gardner opacimeter model Haze-Gard Plus (Labequip, Markham, Canada). Total transmittance is the ratio of transmitted light to the incident light beam. Haze and clarity refer to the loss of contrast due to light diffusion through the film structure, deviating from the incident light beam at angles greater and smaller than 2.5 $^\circ$ C, respectively. The tests were performed in triplicate at room temperature.

2.6. Indoor composting tests

Soil composting tests were carried out according to the method described by Stoll, Silva, Costa, Flôres, and Rios (2017) with modifications. Natural organic soil contained in plastic boxes was used as a composting medium. The film samples (triplicate) were cut (2 cm \times 3 cm), dried at 60 $^\circ$ C to a constant weight (w_0), placed in aluminum meshes (previously dried and weighed) to guarantee contact between the film and soil, and facilitate films collection after a composting time. Subsequently, the film samples were buried at a depth of 5 cm below the composting soil surface. The soil was watered to maintain a moisture level of approximately 40%. The samples were evaluated after 10 days (w_{10}) and their apparent degradation rate was calculated by Eq. (5).

$$\text{Degradation rate (\%)} = \frac{w_0 - w_{10}}{w_0} \times 100 \quad (5)$$

2.7. Statistical analysis

The results were subjected to unidirectional analysis of variance (ANOVA), and the mean values were compared using the Tukey's test at a 95% confidence level ($p < 0.05$), using Origin software, version 6.0 (Origin Lab, Northampton, MA, USA).

3. Results and discussion

3.1. Characterization of mango pulps

Table 1 reports the composition of the mango pulps WIB, IB2, and IB3. No significant difference was observed between these three pulps ($p > 0.05$) in relation to soluble solid content. Regarding pH, the WIB pulp (4.93) was significantly more acidic than the IB3 pulp (5.75) ($p < 0.05$). The highest pH value was observed for the pulp with the highest IB level,

Table 1
Physicochemical composition of mango pulps without (WIB), level 2 (IB2) and level 3 (IB3) internal breakdown (IB).

Sample	Soluble solid content ($^\circ$ Brix)	pH	Fiber content (%)
WIB	17.32 \pm 2.34 ^a	4.93 \pm 0.45 ^b	17.14 \pm 1.88 ^a
IB2	18.08 \pm 1.11 ^a	5.40 \pm 0.33 ^{ab}	12.54 \pm 2.18 ^{ab}
IB3	16.84 \pm 1.43 ^a	5.75 \pm 0.13 ^a	11.55 \pm 2.10 ^b

Mean \pm standard deviation. Values in the same column bearing different letters are significantly different ($p < 0.05$) as per the Tukey's test.

IB3 (5.75), which may relate to the accentuated ripening of mangoes affected by the highest IB level (IB3). The mango ripeness degree has been found to be proportional to the fruit acidity content, increasing the pulp pH value.

For the fiber content, a significant difference ($p < 0.05$) was also observed between the WIB (17.14) and IB3 (11.55) pulps. This result corroborates with those found in the literature (Raymond et al., 1998; Singh, Ram, & Yadava, 2013; Wainwright & Burbage, 1989). According to Malo and Campbell (1978), mangoes with high fiber content are almost unaffected by IB, while genetically improved mango varieties, which have low fiber contents, such as the ‘Palmer’ variety, are more susceptible to this physiological disorder. Thus, mango pulps affected by the most advanced IB stage had a lower fiber content than the pulp extracted from healthy fruits. The middle lamella of the plant cell wall is composed of pectins that contribute to gelation, emulsion stability, and distribution of nutritional fibers (Thakur, Singh, Handa, & Rao, 1997). Pulps from fruits affected by IB tend to show decreased calcium (Ca) content, which results in impaired membrane integrity and cell wall stability (Amarante et al., 2013; Brunetto, Melo, Toselli, Quartieri, & Tagliavini, 2015; Freitas, Amarante, Labavitch, & Mitcham, 2010; Tagliavini, Scandellari, & Toselli, 2016).

3.2. Characterization of collapsed mango-based edible films

The images of the pure pectin (control), WIB, IB2 and IB3 films are shown in Fig. 3.

3.2.1. Morphological analysis

Fig. 4 shows the micrographs of the cross-sectional surface of the control, WIB, IB2 and IB3 films. The control film displayed a smooth and homogeneous microstructure, as the matrix is solely composed of pectin (Fig. 4A). In comparison, the collapsed mango films (Fig. 4B–D) presented a thicker and rougher structure, as previously found for pectin/tare gum edible films (Chen et al., 2020). This characteristic may be associated with the presence of fibers in the mango pulp (Table 1), as observed in edible films from fruit and vegetable residues enriched with pectin (Brito, Carrajola, Gonçalves, Martelli-Tosi, & Ferreira, 2019). The SEM micrographs revealed that film with cohesive matrix with no substantial processing defects were obtained by continuous solution casting, regardless of the IB level.

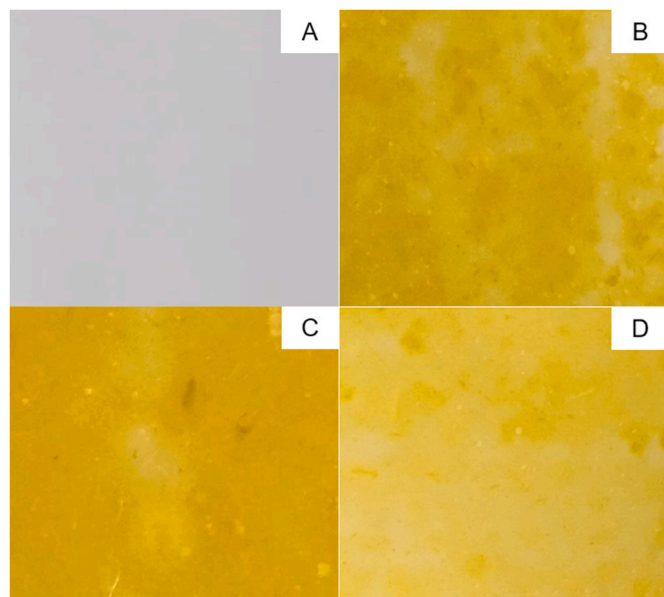


Fig. 3. Images of pure pectin (A), WIB (B), IB2 (C) and IB3 (D) films.

3.2.2. X-ray diffraction (XRD)

XRD analyses were carried out to determine the structure (amorphous or crystalline) of the mango films (Fig. 5). The XRD pattern of pure pectin (Fig. 5D) powder exhibited no reflections, which denotes the amorphous state of pectin (Moreira, De Camargo, Marconcini, & Matoso, 2013). The mango films (Fig. 5A, B and C) present reflections at 17.0° , 19.1° and 22.2° of 2θ , probably related to the starch content in the mango pulp (Oliveira et al., 2018). Therefore, regardless of the IB level, the mango films exhibit a semicrystalline structure, in agreement with the film precursors (mango pulp and pectin).

3.2.3. Spectroscopic characterization by FTIR

The pure pectin film and those containing mango pulp with different IB levels were investigated by ATR-FTIR (Fig. 6). The spectra of WIB, IB2, IB3 and pure pectin (Fig. 6A, B, C and D) exhibited typical vibration bands from 3600 to 3000 cm^{-1} associated with O–H stretching of hydroxyl groups from water molecules and pectin (Singthong, Cui, Ningsanond, & Douglas Goff, 2004). Similar findings were also found in pectin-based films with lemon residues (Dash, Ali, Das, & Mohanta, 2019). It is seen bands between 3000 and 2800 cm^{-1} corresponding to symmetric and asymmetric C–H stretching, also observed by Sucheta, Rai, Chaturvedi, and Yadav (2019). The bands at $1850 - 1630\text{ cm}^{-1}$ correspond to the stretching vibration of carbonyl group (C=O), as also observed in films based on cellulose and pectin added with fruit puree (Viana et al., 2018). The bands between 1200 and 900 cm^{-1} correspond to C–O–H bending vibrations in carbohydrates (Nataraj, Schomäcker, Kraume, Mishra, & Drews, 2008; Shen & Wu, 2003; Wang, Ahmed, Feng, Li, & Song, 2008). These bands comprehend the “fingerprint region” of polysaccharides (Szymanska-Chargot & Zdunek, 2013). The bands at 1045 and 1076 cm^{-1} are also associated with sugars such as xylose, arabinose, and galactose. In general, the spectra were similar, except for a slight increase in some band intensities. This indicates no significant reaction between pectin and mango pulp components during the continuous casting process.

3.2.4. Physical properties of collapsed mango pulp-based films

Table 2 presents the thickness, moisture content, WVP and mechanical properties of the pure pectin, WIB, IB2 and IB3 films. The thickness values varied from 0.08 to 0.28 mm , showing a significant difference ($p < 0.05$) between the mango films and the control. The thickness increased in the order: WIB = IB2 > IB3 > pure pectin. This increase may be associated with the presence of sugars in the mango pulp, which cause a plasticizing effect in the films (Mchugh, Huxsoll, & Krochta, 1996). Among the mango films, WIB and IB2 showed significantly greater thicknesses when compared to IB3. This behavior may be related to the higher fiber content of mango pulps (Table 1). A similar behavior was reported by Liu, Lin, Lopez-Sanchez, and Yang (2020), in bacterial cellulose nanofiber-reinforced edible films, whose thickness increased with the increasing nanofiber content. The thickness of mango film found herein were higher than those found by Martelli, Barros, Moura, Mattoso, and Assis (2013) for films based on ripe banana puree with addition of chitosan, ranging from 0.12 to 0.20 mm , as well as for pectin films with yellow passion fruit residues, ranging from 0.07 to 0.17 mm (Munhoz et al., 2018). In addition, the moisture content of the films ranged from 0.22 to 6.36% and there was a significant difference between the mango films and the control ($p < 0.05$). Similar results were found for puree mango films with a moisture content of 7.96% (Sothornvit & Roodsamran, 2008).

The WVP values of the film samples were significantly different ($p < 0.05$) ranging from 0.13 to $1.54\text{ g mm kPa}^{-1}\text{ h}^{-1}\text{ m}^{-2}$, with a significant increase in WIB (1.18), IB2 (1.54) and IB3 (1.03) compared to the control (0.13). The main polysaccharides present in fruit pulps are pectic and cellulosic substances, in addition, different low molecular weight sugars, with hydrophilic nature, acts as plasticizing agents in the films (Mchugh et al., 1996). The WVP of the mango pulp films was lower than those of açai pulp- (Espitia et al., 2014), apple- (Ravishankar et al., 2012;

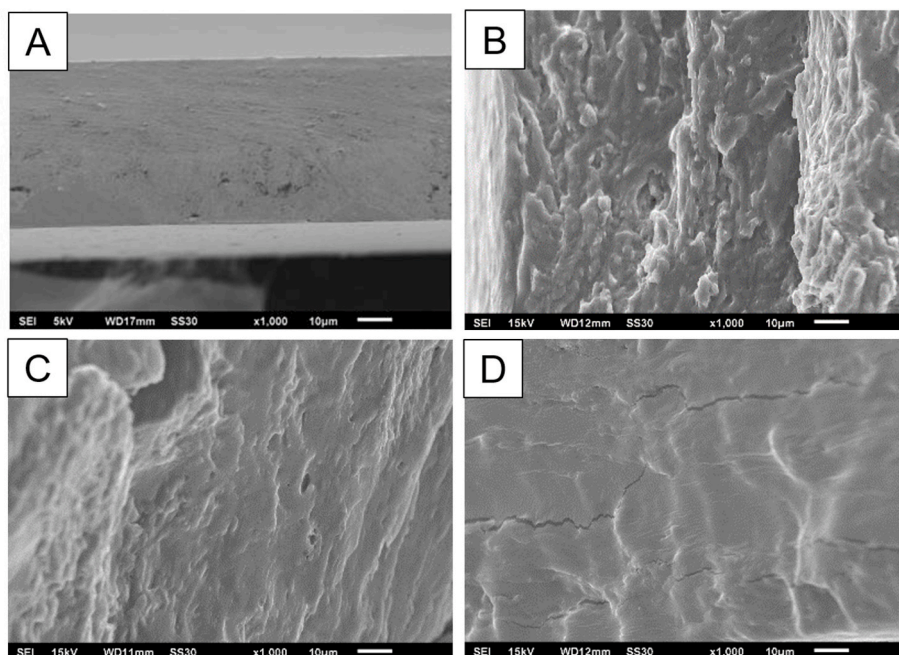


Fig. 4. Scanning electron microscopy (SEM) micrographs of cross-sectional surface for pure pectin film (control) (A), WIB mango films (B), IB2 mango film (C), IB3 mango film (D) at magnification of $1000 \times$. The scale bar corresponds to $10 \mu\text{m}$.

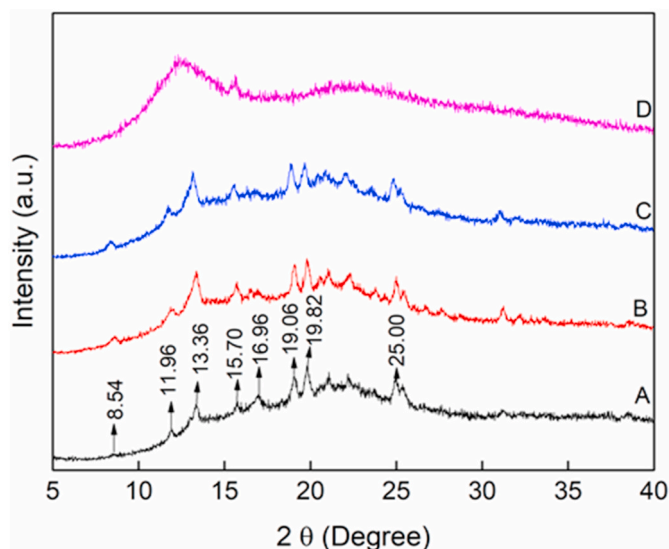


Fig. 5. XRD patterns of WIB (A), IB2 (B) and IB3 (3), and pure pectin (control) (D) films.

Ravishankar, Zhu, Olsen, McHugh, & Friedman, 2009), papaya- (Lorveice, Moura, Aouada, & Mattoso, 2012) and tomato-based films (Otoni et al., 2014). Therefore, it is possible to suggest that the mango films (WIB, IB2 and IB3) are within the same range of water barrier properties for food packaging applications.

Mechanical properties are key requirements of packaging films. Low mechanical strength or ductility hinder film functionality in the production, handling, and storage stages (Shafie, Yusof, Samsudin, & Gan, 2020). The tensile test results showed that the pure pectin film (control) had significantly ($p < 0.05$) greater tensile strength and lower elongation at break when compared to the mango pulp films. This behavior can be explained by the plasticizing effect of the sugars present in the mango pulp. The pectin matrix free of plasticizers encompasses polymer chains with less mobility, reducing the ductility and providing large

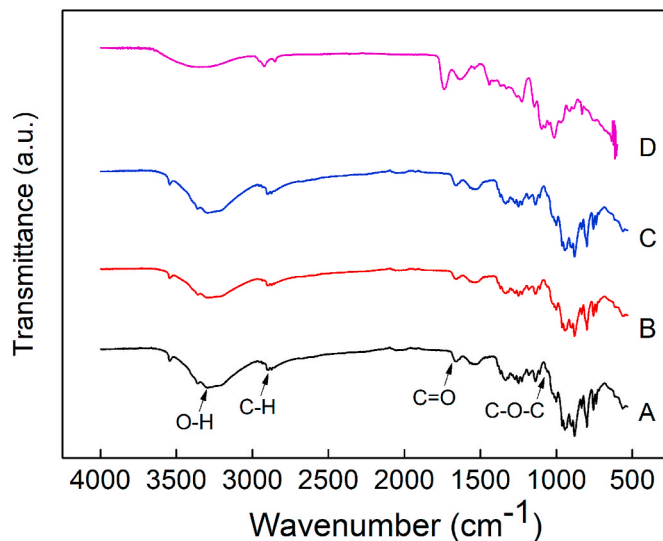


Fig. 6. ATR-FTIR spectra of WIB (A), IB2 (B), IB3 (C) and pure pectin (D) films.

mechanical resistance to the film (Shafie & Gan, 2020).

The significant difference in tensile strength indicates that the IB level influenced the mechanical resistance of the films. The tensile strength of WIB was 7.64 MPa, significantly higher than the values found for IB2 and IB3, 3.40 and 3.38 MPa, respectively. This may be due to the lower fiber content found in the collapsed pulps (Table 1), since fibers have a reinforcing effect on polymer matrices.

Films loaded with fibers have good mechanical properties, which are related to the good adhesion at the polymer-fiber interface, which promotes an effective transfer of stress from the polymeric matrix to the fiber (Kuciel, Mazur, & Jakubowska, 2019). The increase in tensile strength with increasing fiber content of fruit puree-based films has been reported in other studies (Kuciel et al., 2019; Ma et al., 2020; Varghese, Pulikkalparambi, Rangappa, Siengchin, & Parameswaranpillai, 2020).

The elongation at break values (Table 2) increased significantly in

Table 2

Thickness (mm), moisture content (%), water vapor permeability (WVP), and mechanical properties (tensile strength (TS) and elongation at break (ϵ)) of films based on pure pectin and mango without (WIB), level 2 (IB2) and level 3 (IB3) internal breakdown (IB).

Filmes	Thickness (mm)	Moisture content (%)	WVP (g mm kPa ⁻¹ h ⁻¹ m ⁻²)	TS (MPa)	ϵ (%)
Pectin	0.08 ± 0.004 ^c	0.22 ± 0.17 ^b	0.13 ± 0.07 ^c	35.57 ± 2.81 ^a	4.27 ± 1.08 ^c
WIB	0.28 ± 0.01 ^a	6.36 ± 1.14 ^a	1.18 ± 0.02 ^b	7.64 ± 1.60 ^b	25.73 ± 6.71 ^b
IB2	0.29 ± 0.03 ^a	5.66 ± 0.55 ^a	1.54 ± 0.09 ^a	3.40 ± 0.57 ^c	72.63 ± 7.57 ^a
IB3	0.23 ± 0.02 ^b	5.98 ± 0.31 ^a	1.03 ± 0.04 ^b	3.38 ± 0.71 ^c	62.55 ± 17.32 ^a

Mean ± standard deviation. Values in the same column bearing different letters are significantly different ($p < 0.05$) as per the Tukey's test.

the films composed of collapsed mango pulp (IB2 and IB3), corroborating with the plasticizing effect of the mango pulp sugars (Munhoz et al., 2018; Viana et al., 2018). The high sugar content in the corresponding pulps relates to the IB stage because the collapse accelerates mango ripening (Seshadri, Manoharan, & Singh, 2019), making the ensuing films more flexible.

3.2.5. Optical properties

Optical properties are important parameters of food packaging materials, as they relate to the acceptance of food product by consumers (Song, Zuo, & Chen, 2018). Table 3 presents the color analysis results for the pure pectin, WIB, IB2 and IB3 films.

As observed in Table 3, the WIB and IB2 films showed a slightly darker color than the pure pectin film, based on the L* (brightness) values, 80.6 and 78.7, respectively. L* values above 70 represent films with light colors, which is recurrent in edible films (Basiak, Debeaufort, & Lenart, 2016; Galus & Kadzi, 2016). Regarding the chromatic coordinates, the a* values ranged from -0.2 to 3.5 and a significant difference was observed between IB2 and IB3 ($p < 0.05$). The IB3 film was slightly greenish, while the IB2 film showed a slightly reddish color based on its positive a* value. For the b* values, a significant variation from -7.8 to 68.6 was observed between the mango films and the control ($p < 0.05$). The mango pulp provided color to the films, which is evidenced by the increase in the yellowish tone (positive b* coordinate values) ranging from 60.2 to 68.6 ($p < 0.05$). Munhoz et al. (2018) also observed an increase in the yellowish color of films based on passion fruit pulp, as well as in films with a 50/50 pectin/passion fruit peel ratio. Positive a* and b* coordinates imply in predominant reddish/yellowish coloration in the films, respectively. The a* value may be related to the predominance of carotenoids and lycopene, and the parameter b* relates to the orange color of mango fruits due to β -carotene (Pathare et al., 2013).

The C* values of the films ranged from 7.9 to 68.7, with greatest saturation for the IB2 film and the lowest value for the control ($p < 0.05$). The increase in C* is indicative of chlorophyll degradation and carotenoid synthesis, which reflects an increase in the yellowish color intensity observed in the mango films (Lawless & Heymann, 2010).

Table 3

Colorimetric parameters of luminosity (L*), a*, b* chroma (C*), hue angle (h°), total color difference (ΔE^*), transmittance (%), haze (%) and clarity (%) of pure pectin, WIB, IB2 and IB3 films.

Films	L*	a*	b*	C*	h°	ΔE^*	Transmittance (%)	Haze (%)	Clarity (%)
Pectin	89.8 ± 0.2 ^a	1.1 ± 0.02 ^{ab}	-7.8 ± 0.03 ^d	7.9 ± 0.04 ^d	278.1 ± 0.2 ^a	7.8 ± 0.0 ^d	87.9 ± 0.7 ^a	7.0 ± 0.7 ^d	96.9 ± 0.1 ^a
WIB	80.6 ± 1.3 ^b	2.0 ± 1.1 ^{ab}	64.4 ± 1.6 ^b	64.4 ± 1.6 ^b	88.3 ± 0.9 ^b	65.8 ± 1.8 ^b	71.7 ± 0.4 ^c	77.9 ± 0.3 ^b	13.1 ± 0.2 ^b
IB2	78.7 ± 2.0 ^c	3.5 ± 1.7 ^a	68.6 ± 1.7 ^a	68.7 ± 1.8 ^a	87.1 ± 1.4 ^c	70.3 ± 1.7 ^a	66.7 ± 0.5 ^d	82.2 ± 0.3 ^a	12.1 ± 0.1 ^c
IB3	83.6 ± 0.3 ^{ab}	-0.2 ± 0.3 ^b	60.2 ± 0.9 ^c	60.2 ± 0.9 ^c	90.2 ± 0.3 ^{ab}	61.1 ± 0.9 ^c	77.5 ± 0.3 ^b	74.5 ± 0.2 ^c	13.4 ± 0.1 ^b

Mean ± standard deviation. Values in the same column bearing different letters are significantly different ($p < 0.05$) as per the Tukey's test.

The h° values, which defines red at 0°, yellow at 90°, green at 180°, and blue at 270°, ranged from 87.1 to 278.1, indicating that the films had a predominance of yellow tint, except for IB3, whose h° value between the yellowish and bluish interpretation, when compared to the pure pectin film. The total color difference (ΔE^*) ranged from 7.8 to 70.3 and significant differences are observed between all samples ($p < 0.05$), demonstrating that the films had different colors distinguishable with the naked eye. Thus, the mango films were clear (L* value closer to 100), with yellow (h° close to 90) and more saturated (C* more distant from zero) tones.

The light barrier properties are important properties of packaging films in terms of food protection against deterioration caused by visible and ultraviolet light, since photosensitive foods can suffer oxidation, loss of nutrients, and develop unpleasant flavors (Crizel et al., 2018; Han, Yu, & Wang, 2018). Luminous transmittance (%) represents the total percentage of incident light that is transmitted through the packaging material. Films with transmittance above 90% are considered as transparent structures (Hernandez, 1997). The transmittance values ranged from 66.7 to 87.9% ($p < 0.05$), the haze values ranged from 7.0 to 82.2% ($p < 0.05$), whereas the clarity values ranged from 12.1 to 96.9%, with significant differences ($p < 0.05$) between the mango films and the control, the latter being the clearest sample.

Thus, the mango films with different IB levels were opaque and more colorful, with tones ranging from light yellow to dark, which is attributed to the pulp chemical composition at the different IB levels, particularly the presence of pigments (carotenoids and lycopene, β -carotene related to the orange color of mango fruits) (Solovchenko, Yahia, & Chen, 2019). Given these characteristics, the collapsed mango films developed herein can be suitable as packaging of light-sensitive foods (Crizel et al., 2018).

3.2.6. Biodegradability tests

To gain a preliminary insight into the biodegradability of the collapsed mango pulp-based films, a qualitative soil composting study was performed using indoor conditions. The film weight could not be determined precisely because the soil impregnated the sample after they were removed from the composting medium. The images of the films after the test are shown in Fig. 7.

It can be seen that the coloration of all films changed after the composting test. The films disintegrated almost entirely over the 10th day of incubation. Overall, the components of the mango pulp are easily assimilated by microorganisms (Cinelli et al., 2014; Medina Jaramillo, Gutiérrez, Goyanes, Bernal, & Famá, 2016). IB-affected mango pulps tend to show a low calcium (Ca) concentration. This impairs the maintenance of the membrane integrity and stability of the cell wall (Amarante et al., 2013; Freitas et al., 2010; Tagliavini et al., 2016), favoring the decomposition of mango pulp macromolecules. The degradation of pectic substances follows two routes: (i) depolymerization and (ii) demethylation. The first is associated with β -elimination reactions stimulated by pectin lyases from microorganisms, and the second occurs with the aid of pectin esterase, followed by acid hydrolysis of α (1 → 4) glycosidic bonds by polygalacturonases (Singthong et al., 2004).

Considering the short disintegration time (10 days), it is possible to suggest that the films obtained from IB-affected mango pulps can be composted, favoring a rapid mineralization by the soil microbiota and

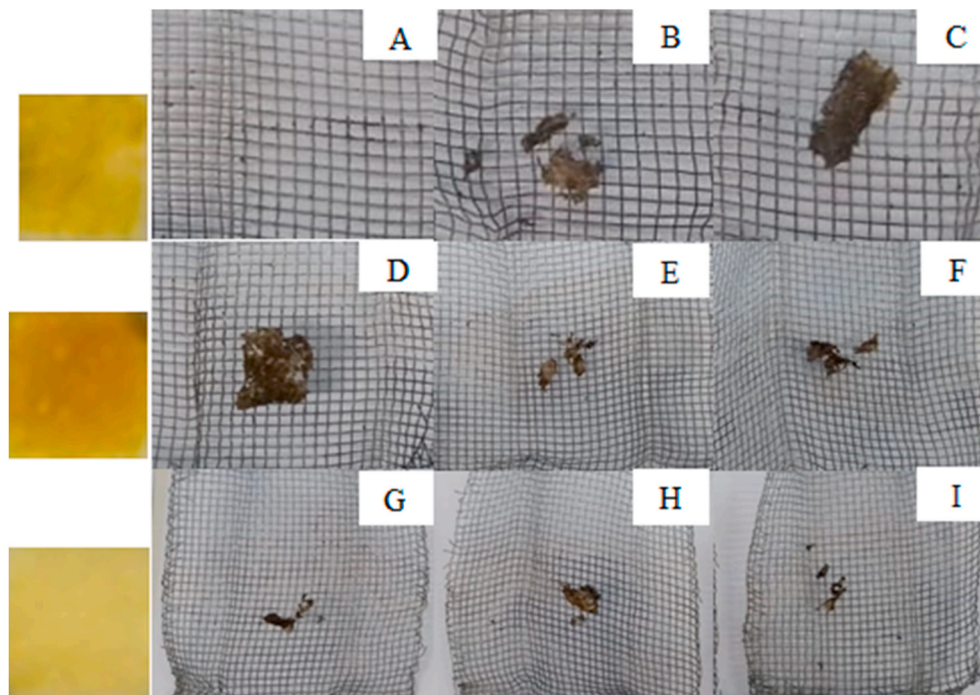


Fig. 7. Image of WIB (A, B and C), IB2 (D, E and F) and IB3 (G, H and I) after 10 days of composting test.

the metabolization of biopolymers, converting them into elementary substances (carbon dioxide, methane, and water) and biomass (Mohee, Unmar, Mudhoo, & Khadoo, 2008). Therefore, the production of edible films from IB-affected mangoes becomes an alternative to add value to fruits that could potentially be rejected by consumers, while it is a sustainable approach to produce films that could replace petroleum-derived materials, thus reducing the environmental impact caused by non-biodegradable plastic packaging.

4. Conclusion

Mangoes pulps affected by IB were successfully turned into edible films by continuous solution casting. This technique allowed for good retention of the natural mango pulp coloration in the films, while increasing the edible film productivity. The edible film properties were found to be affected by the IB level, as the physicochemical parameters of the mango pulps changed with IB progression. Films with highest water vapor permeability, and largest ductility and opacity were obtained from mango pulps affected by the most advanced IB stage. Overall, injured mango pulps can be used as raw materials to produce edible films with suitable properties for food packaging applications. In a broader perspective, the continuous casting approach described herein can be applied to other injured fruits as a means of valorizing fruits that are typically wasted due to aesthetical imperfections.

CRedit authorship contribution statement

Fernanda C.A. Oldoni: Conceptualization, Methodology, Investigation, Data curation, Writing – original draft. **Marcela P. Bernardo:** Methodology, Writing – original draft. **Josemar G. Oliveira Filho:** Methodology, Writing – original draft. **Aline C. de Aguiar:** Methodology, Investigation. **Francys K.V. Moreira:** Supervision, Writing – review & editing. **Luiz H.C. Mattoso:** Supervision, Writing – review & editing. **Luiz A. Colnago:** Supervision, Writing – review & editing. **Marcos D. Ferreira:** Conceptualization, Funding acquisition, Supervision, Writing – review & editing.

Declaration of competing interest

The authors declare that there is no conflict of interests regarding the publication of this paper.

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