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Development and evaluation of a biodegradable packaging from the aryl of the fruit of Hymenaea stigonocarpa Mart. ex Hayne

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Abstract. The fruits of *Hymenaea stigonocarpa* have a sweet, starchy fibrous structure used as food among wild animals and man in the field. The study aimed to develop and evaluate a biodegradable packaging from the starchy aryl solution of the jatobá fruit (*H. stigonocarpa*). The amylaceous solution was obtained from the arils of *H. stigonocarpa*, the biodegradable film was prepared according to the casting technique. The physical-chemical characteristics for thickness (mm), humidity (%), water solubility (%), biodegradability time and transmittance (T%) were evaluated. Morphology by optical micrographic and scanning electron, mathematical modeling in 3D, and for the mechanical parameters of tensile strength, maximum tension, elongation and elasticity module. The results obtained for the biodegradable film were brown yellow color, aroma and homogeneity, thickness 0.27 mm, humidity 12.45%, solubility in water 57.48%, biodegradability of 100%, maximum and minimum transmittance 82.25 and 1.32 (T%), in the optical micrograph small imperfections were observed and in the scanning electron micrograph small cracks. The mathematical modeling in 3D presented a surprising result, which is an important device in the aid of imaging. The mechanical characteristics for maximum tension 3.17 N, rupture stress 1.34 MPa, elongation 2.99% and elasticity with 90.07 MPa presented satisfactory results comparable to other biodegradable films of native starch. Biodegradable packaging from *Hymenaea stigonocarpa* aryl has great potential for use as a food packaging.
Keywords: Biodegradable film, *Hymenaea* Genus, Food packaging, Natural packaging

Introduction

Hymenaea stigonocarpa Mart. ex Hayne, belongs to the genus *Hymenaea*, popularly known as "*jatobá-do-cerrado*" has a homogeneous distribution, occurring in tropical regions, mainly in areas of the Brazilian Cerrado domain (Paiva; Machado, 2006; Lacerda; Malaquias; Peron, 2014).

This species of the Fabaceae family: Caesalpinioidae, produces annual crops with large fruits in the form of dry, indehiscent, monospérmic or polyspermic vegetables, elongated, rounded and slightly rectified apex, the rounded base and the entire margin slightly wavy, with a diameter between 8.7-20.0 cm long, with a light brown to dark brown color, inside there is the yellow, aromatic, fibrous-floury and sweet aryl, rich in starch that covers the seeds (Botelho et al., 2000; Menezes Filho et al. , 2019).

Due to this characteristic, the aryl, being constituted by a certain percentage of starch, becomes a possible candidate for the development of biodegradable packaging.

Currently, divers natural polymer matrices are known, mainly based on starch, chitosan, gelatin, fish myofibrillaries, corn zein, wheat gluten, collagen, polysaccharides, lipids and proteins, where, in several studies, they incorporate essential oils, oil-resin, fixed oils, among other compounds such as plant extracts and active substances such as lycopene, beta-carotene, among others, promoting this product, antioxidant and antifungal characteristics, glycerol is also incorporated, which aims to promote in the polymer matrix an organized arrangement acting as plasticizers (Kechichian et al., 2010; Assis et al., 2017; Caetano et al., 2018; Malherbi et al., 2019; Fuente et al., 2019; Marasca; Nogueira; Martins, 2020).

Biodegradable starch films are inexpensive, easily available and have good physicochemical, morphostructural and mechanical characteristics (barrier to external and internal elements), guaranteeing their use in various activities, mainly for maintenance, avoiding contamination and preserving characteristics of foods that you want to preserve for a longer shelf life (Henrique; Cereda; Sarmento, 2008; López et al., 2011; Ezeoha; Ezenwanne, 2013; Lucena et al., 2017; Silva et al., 2019). These can be extracted directly from vegetables, a was as obtained by hydrolysis processes, known as modified starches (Asiri; Ulbrich; Flöter, 2019; Ulbrich; Daler; Flöter, 2019; Oliveira Filho et al., 2020).

In addition natural packaging, combines environmental preservation and reduces the inappropriate use of synthetic petroleum-based packaging, which presents a long period of decomposition in the environment, being a serious problem in the conservation of aquatic species where they present a greater risk (Nor Adilah et al., 2018; Bernardi; Hermes; Boff, 2018; Menezes Filho; Sousa; Castro, 2020).

The objective this study was to develop and evaluate a biodegradable packaging based on the starch solution obtained from the aryl of the fruit the *Hymenaea stigonocarpa* by the casting process.

Methods

Production of biodegradable packaging (Film)

The film was obtained by a casting technique, with the use of the methodology proposed by Issa et al. (2017), adapted. In order to produce film, 10 g of aryl was dissolved in 250 mL deionized water. The solution was then moderately agitated at room temperature (25 °C) for 30 minutes. The solution was filtered under manual pressure on thin nylon fabric, the supernatant was collected. Afterwards, it was heated at 70 °C, at constant agitation for 30 minutes. After starch solution gelatinization, glycerol (Dinâmica, PA. – ACS, purity of 99,5%) was added as a plasticizer (35% p/p); this dispersion was agitated (BiomiXer, Mod. 78HW-1) for five more minutes. Filmogenic solution was poured on polystyrene (20 cm) (Kasvi, Mod. CRAL) slabs and dried in an air circulation (Ethik, Mod. 40 L) oven at 35 °C for about 36 hours.

Physicochemical characterization

Film thickness

Film thickness was measured by a digital caliper, (Matrix, Mod. 150 mm Mtx) whose precision was 0.01 mm. Measurements were carried out in 15 spots on every film and the thickness mean was calculated (Santos et al., 2020).

Moisture content

The film was weighed and then oven dried at 105 °C (Thoth, Mod. Th-510-480) for 4h. according to the methodology described by Rambabu et al. (2019) adapted.

Measurement of water solubility

Film which measured about (2 x 2 cm) were dried in an oven at 105 °C for 4 hours and, then weighed so that initial mass (*Mi*) could be determined. They were immersed in 60 mL distilled water and kept under constant agitation at 25 °C for 24 hours. Afterwards, solutions with the films were filtered through filter paper (Unifil, Mod. C42) which had been previously weighed. Sheets of filter papers with films were dried at 105 °C for 24 hours and weighed so that final mass (*Mf*) could be found, in agreement with the methodology described by Jahed et al. (2017), adapted. The film solubility (%) was calculated by equation (1).

$$WSol(\%) = (Mi - Mf/Mi)\ x\ 100 \quad \text{Eq. (1)}$$

Where: *Mi* = initial mass and, *Mf* = final mass.

Biodegradability time

The analysis was carried out by the methodology described by Martucci and Ruseckaite (2009), adapted. Film samples (2 x 2 cm) were dried up to constant weight so that initial mass (*Mi*) could be determined. Samples were then placed in open polyethylene packages, high density polyethylene (HDPE) to enable microorganisms and moisture to gain access to them.

After that, they were buried in organic soil, which had been previously prepared, at constant moisture and room temperature. Twenty days after the experiment installment, the packages with the samples were removed from the soil, washed with distilled water and dried up to constant weight (*Mf*). Biodegradability (%) and, moisture (%) was calculated by equation (2).

$$Bio(\%) = (Mf - Mi/Mi)\ x\ 100 \quad \text{Eq. (2)}$$

Where: *Mf* = final mass and, *Mi* = initial mass.

Light transmittance rate (UV-Vis)

Ultraviolet (UV) and visible light transmittance of film was conducted by UV-*Vis* spectrophotometer (Belphotonics, Mod. M-51). Film samples were cut and placed in cuvettes so that transmittance could be measured over a wavelength range between 250 and 850 nm (Hosseini et al., 2015).

Morphology by scanning optical and electron microscopy

The film was fixed on a microscope slide. Then, it was observed under an optical microscope (Nikon, Mod. ED200) with magnifications of 4, 10 and 40 x. Micrographs were taken using a camera attached to the microscope (Nikon, Mod. DS-Ri2).

The film was fixed on brass sample holders. Images were carried out by a JEOL (JSM, Mod. 6610) (EDS, Thermo Scientific NSS Spectral Imaging) scanning electron microscope (SEM) in high-vacuum mode to detect secondary electrons at electron accelerating voltage of 3 kV.

Mathematical modeling

Mathematical modeling was performed using ImageJ software (free version, 1.8.0_172).

Resistance assay

The traction resistance was determined using a universal machine (Instron, Mod. 3360). Film strips were cut with a diameter of (15 x 120 mm) as described by Menezes, Souza and Castro (2019). The system was adjusted with 100 mm hook spacing. The tensile strength was carried out with a speed of 0.21 mm/s, in a maximum load cell of 650 N. The results were obtained for maximum tension, breaking stress, elongation and for modulus of elasticity.

Statistical analysis

Analyses were carried out in triplicate and standard deviations (±) were calculated. Means were obtained with the use of the PAST 3 software program (free version, 2019).

Results and discussion

In Figure 1, the image of the biodegradable film obtained from the aryl of the *Jatobá* fruit (*H. stigonocarpa*).

Figure 1. Biodegradable film of the aryl of fruit of *Hymenaea stigonocarpa*. Source: Authors, 2021.

The biodegradable film showed a brownish yellow visual color, with a slight natural aroma of the fruit aryl. Menezes Filho, Souza and Castro (2019) also obtained biodegradable yellow colored film from the watermelon peel. According to Rocha et al. (2014) the color of biodegradable films can affect their acceptance in both edible and non-edible applications (packaging).

The film was good aspect, easy handling and was visibly homogeneous, with no presence of insoluble particles or brittle areas. Similar results were obtained by Malherbi et al. (2019) with gelatin-based corn starch biodegradable films incorporated with guabiroba pulp.

Table 1, shows the results of the physicochemical and mechanical parameters obtained by the biodegradable aryl film of the fruit of *H. stigonocarpa*.

The thickness obtained in the biodegradable film is within the range observed in other studies from alternative starch material (Tab. 1). The study by Assis et al. (2017) the researchers found for biodegradable films of cassava starch incorporated with lycopene nanocapsules thickness between 0.11-0.15 mm. Souza, Silva and Druzian (2012) found a thickness between 0.12-0.14 mm and a control of 0.12 mm in different formulations of biofilms of cassava starch incorporated with mango and acerola pulp. The according Hosseini et al. (2015), Adilah et al. (2018) and, Santos et al. (2020), film thickness depends on factors related to drying and to the method of preparation. Thickness should be measured because it affects mechanical properties of films, such as water vapor permeability.

Possibly the aryl of the jatobá fruit has a high amylose content, and further studies are needed to determine this natural polymer, as observed in (Tab. 1). This polymer induces high sensitivity to moisture, which may also affect mechanical properties of films (Thakur et al., 2019).

The moisture content of the film, presented similarity to other biodegradable starch based packaging. Assis et al. (2017) reported having found for biodegradable films of cassava starch incorporated with lycopene nanocapsules, moisture content between 10.68 to 13.46%.

The biodegradable film of the *Jatobá* fruit aryl showed a moderate percentage of solubility (Tab. 1), which is, restricted use in environments with low humidity saturation. The lower the percentage of solubility, the better the quality and durability of the biodegradable packaging, being able to stay in an environment with a certain level of relative humidity. Solubility can also be influenced by the pH and glycerol effects, which can interact in the structural conformation of the polymer (Rocha et al., 2014). Solubility is also an important parameter that should be analyzed in starch packaging. The ideal level of film solubility depends on their final use, example: food, transport of liquids and solids. Starch is a hydrophilic material, thus, when a starch film is exposed to water, its polymeric molecules form hydrogen bonds with water and lead to film dissolution (Bertuzzi et al., 2007; Kim et al., 2015).

After the 20-day analysis, the polyethylene packages that contained film of the aryl of fruit the *H. stigonocarpa* was removed from the soil. Since film had been thoroughly degraded, the material could not be weighed to quantify biodegradability. This is a good result because which may be considered a promising material for biodegradable and eco-friendly packaging. The same was observed in the study by Santos et al. (2020), where they used different filmogenic solutions from arrowroot starch. The researchers at time of 30-days, observed the packages were completely degraded, being impossible to determine the biodegradability. The according of Martucci & Ruseckaite (2009), and Assis et al. (2017) biodegradable analysis of the film in a *in natura* soilseeks, to play a process of degradation in natural environment. Where the soil is generally comprised of microflora consists of bacteria, fungi and protozoa which act in synergy in the biodegradability process in less time.

As similar results were observed was observed by Medina Jaramillo et al. (2016) in cassava starch films with yerba mate extract, and Seligra et al. (2016) in biodegradable films of starch with rapid biodegradability of the material in 12-15 days respectively, where showed a significant degradation in 30-days, demonstrating the importance of development biodegradable packaging and substitution packaging obtained from non-biodegradable polymers (petroleum-based synthetic).

According of Maran et al. (2014), Seligra et al., 2016, and Assis et al. (2017) biodegradable films obtained from native starch and glycerol, which compounds exhibit hydrophilic character, can present high loss of mass during in the biodegradation, due to increased water absorption by starch. The increased water absorption of polymers promotes the growth of microorganisms naturally present in the soil, that act on the rich source of carbohydrate and results in a greater and more rapid degradation of these compound.

In Figure 2, the transmittance rate of the biodegradable film of the aryl of fruit of *H. stigonocarpa*.

Table 1. Physicochemical and mechanical parameters of the biodegradable film of the *Jatobá* (*Hymenaea stigonocarpa*) fruit.

Parameters	**Results***
Thickness (mm)	0.27 ± 0.11
Moisture content (%)	12.45 ± 0.92
Water solubility (%)	57.84 ± 4.11
Biodegradability (%)	100 ± 0.00

*Average of the triplicate followed by (±) standard deviation. Source: Authors, 2021.

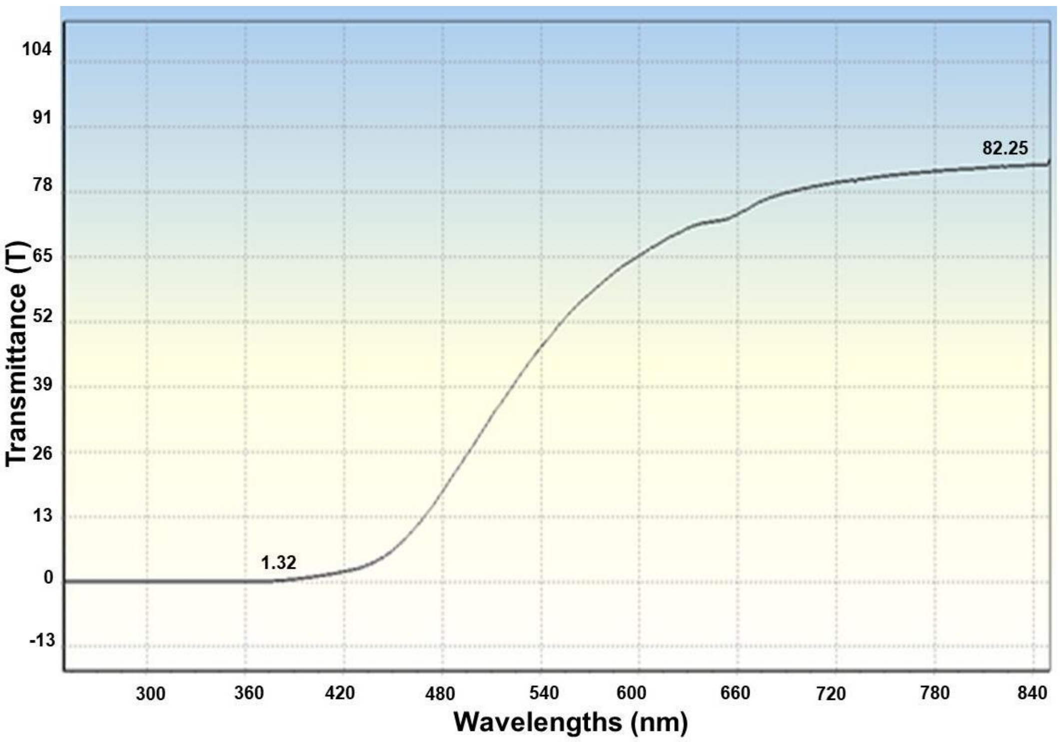

Figure 2. UV-*Vis* light transmittance rate in biodegradable film of aryl of the fruit of Jatobá (*Hymenaea stigonocarpa*. Source: Authors, 2021.

The film showed a similar behavior when subjected to the analysis of UV and visible light transmission at different wavelengths, from 250 nm to 850 nm with other studies. The maximum transmission was 82.25% and the minimum 1.32% as seen in (Fig. 2). A similar result was observed in the work of Assis et al. (2017) in cassava starch films with lycopene nanocapsules with maximum transmittance from 86.56% (800 nm) to 5.19% (200 nm).

A color is an important parameter to be evaluated since it influences consumers acceptance in the products (Lynch; Kastner; Kropf, 1986; Magnier; Schoormans, 2017). According of Romani et al., (2018), Rambabu et al. (2019), and Santos et al. (2020) films used as traditional packaging are usually transparent so that the product can be seen more easily. Transparent packaging is more attractive from the consumer's point of view, however, colorful and opaque biodegradable films help to protect food exposed to visible and UV light, mainly in food with high fat content, preventing them from undergoing oxidative degradation negatively influencing in the quality.

In Figure 3, we can observe the images by optical micrographic, electronic scanning and 3D mathematical modeling of the surface area of the biodegradable aryl film of *H. stigonocarpa* fruit.

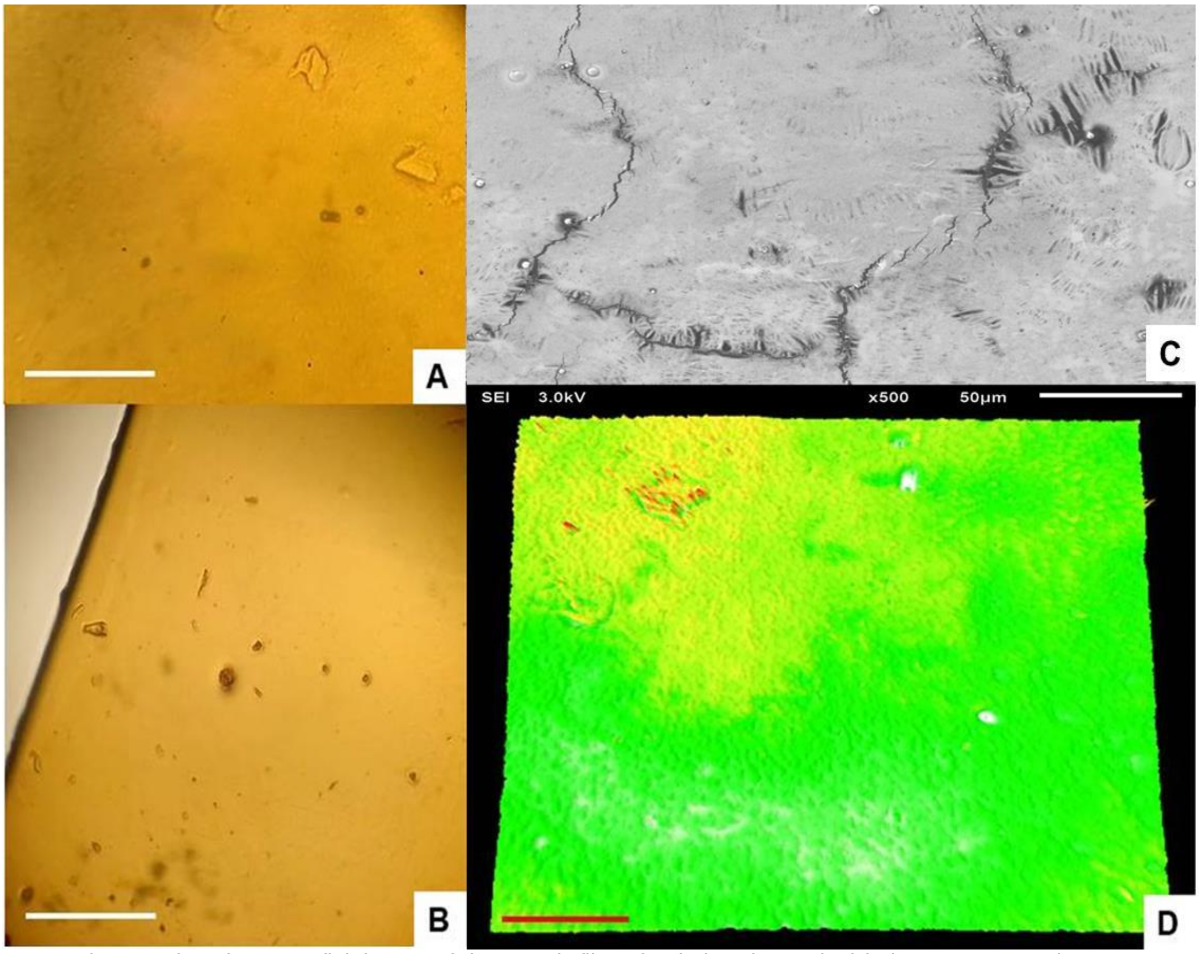

Figure 3. Micrographs of a superficial area of the starch film of solution the aryl of fruit *Hymenaea stigonocarpa*. Bars: (**A**) 65 µm, (**B**) 80 µm, (**C**) 50 µm, (**D**) 90 µm. Source: Authors, 2021.

The biodegradable film of aryl *Jatobá*, presents good malleability, good appearance and aroma. Brito et al. (2019) evaluated biodegradable films of flours and pectin from the residues of orange, passion fruit and watermelon, where they found similar results to this study (Fig. 3, lattes A, B and D), without roughness, with a shiny appearance, good malleability and, fruity aroma. In the scanning electron micrograph, it is possible to verify a surface area with cracks, probably related to the drying process, in addition to small starch granules (Fig. 3, letter C).

In the 3D mathematical model, one can observe the moderately uniform topography of an area of the surface observed on both faces (Fig. 3, last D). In green, homogeneous surface area, yellow area with small elevations and in red substantial elevations. The white dots represent the starch granules. The results for maximum stress (N) = 3.17 ± 0.12, rupture stress (MPa) = 1.34 ± 0.66, elongation (%) = 2.99 ± 0.76, elasticity (MPa) = 90.07 ± 14.21.

They are similar to those observed by Menezes Filho, Souza and Castro (2019) for biodegradable watermelon film, with maximum tension of 1.38 MPa, elongation of 2.26%, elasticity module of 91.09 MPa and breaking strength of 3.01 N. Malherbi et al. (2019) found results between 10.95-79.04 MPa for tensile strength and 1.45-6.19% rupture elongation for biodegradable corn starch films incorporated with guabiroba pulp. Already Assis et al. (2017) obtained results of tensile strength between 2.43 to 18.13 MPa and elongation at break between 61.27 to 399.92% for biodegradable films of cassava starch incorporated with lycopene.

Conclusion

The development of biodegradable film from renewable sources, as starch in the aryl of the fruit *Hymenaea stigonocarpa*, may be an alternative to non-biodegradable packaging, with high biodegradability in a short period of time. The biodegradable film from the aryl of the *Jatobá* fruit, presented interesting results aiming at the easiness of obtaining the raw material and of production. The results of the physicochemical, morphological and mechanical parameters qualify this new product for the packaging production market as a possible substitute for synthetic packaging used of foods.

This is first of several studies on the development of new biodegradable packaging that seeks to combine the preservation of an endemic species, in the Cerrado with the production of green line packaging for sustainable products.

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