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Edible Films and Coatings Formulated with Arrowroot Starch as a Non-Conventional Starch Source for Plums Packaging

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Abstract: Increasing environmental awareness has promoted an interest in alternative strategies to common plastics obtained from fossil sources, stimulating research on the use of biodegradable and edible films/coatings obtained from renewable sources such as arrowroot starch. This research work aimed to evaluate the use of arrowroot starch on the formation of edible films and coatings. Increasing the concentration of arrowroot starch (from 1% to 5%, mass/mass) in the film produced by casting resulted in increased water vapor permeability (from 2.20 to 3.68 g mm/m^2 day kPa), moisture content (3.22% to 7.95%), increased thickness (from 0.029 to 0.101 mm), and decreased solubility in water (from 22.45% to 13.89%). The films were homogeneous, transparent and manageable, with the exception of the film with 1% starch. Film-forming solutions at concentrations of 0%, 2%, and 4% (mass/mass) of arrowroot starch were prepared and applied to plums to evaluate post-harvest behavior when stored at 25 and 5 °C for 35 days. The 2% coating adhered well to the plums' surfaces, was bright and was effective in reducing mass loss and respiratory rate, associated with storage temperature of 5 °C. The 4% coating presented an opaque and flocculated appearance.

Keywords: *Maranta arundinacea* Linn.; *Prunus domestica*; food packaging; biopolymers; solubility in water; post-harvest technology; anthocyanins; cooling

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1. Introduction

Concerns about the environment combined with consumer demand for high quality, environmentally friendly, closer to natural products, have drawn researchers' attention to the development of technologies that replace the consumption of fossil materials with sustainable processes and materials from renewable sources [1]. In recent decades, new packaging technologies have been developed in response to this demand, and one of the found solutions was the development of biodegradable and edible films and coatings from natural polymers from renewable sources [2].

The biopolymers most often used in the production of edible films and coatings are proteins, polysaccharides and lipids, or the combination thereof [3]. Among the polysaccharides used for edibles films and coatings production, starch is the most widely used natural biopolymer, due to its easy processing, low cost, abundance, biodegradability, edibility and ability to form a continuous matrix [4]. Starch is a vegetable energy reserve polysaccharide, formed by two types of glucose polymers, amylose and amylopectin, with different structures and functionalities, whose composition (amylose/amylopectin ratio) varies according to the biological source. Amylose is a linear polymer, while amylopectin

is a highly branched and less water-soluble polymer than amylose [5]. Starch is extracted commercially from several sources, such as corn, potatoes, cassava, rice and wheat [6]. Its wide use in the food industry, in several types of foods as gelling and thickener, has intensified the search for new natural sources of unconventional starch from yam, jackfruit seed, mango kernel [7], avocado [8], mango ginger [9] and arrowroot [10] among others.

In this sense, arrowroot rhizomes (*Maranta arundinacea* Linn.) have stood out, due to their high starch content and because they are a starch source of no socioeconomic importance in many countries and, therefore, are not considered a high priority raw material [11]. The high content of amylose (ranging from 16% to 27%, [12]) makes arrowroot starch a promising polymeric material to be used in biodegradable films preparation [10]. The high amylose content in the starch is desirable for the production of films with better mechanical and thermal properties [13–15]. In addition, the high digestibility of arrowroot starch [16] can also be considered an advantage for applications in which the films or coatings will be consumed together with the food contained. Nowadays, one of the alternatives to maintain or improve the quality in the post-harvest of fruit and vegetables is the application of an edible coating or film to the surface of the product.

The film is performed separately and applied subsequently to the fruit and vegetables [17]. Edible coatings are applied to or formed directly over the surface of fruit and vegetables, creating thin membranes, imperceptible to the naked eye and with several structural characteristics, which are dependent on the formulation of the precursor film-forming solution [18]. As they are in direct contact with food and can be consumed with food, it is desirable that edible films and coatings have neutral sensory properties (transparent, odorless and tasteless) so as not to alter food quality [3]. The purpose of the edible coatings is to create a selective barrier to gases, aromas and oils to improve the food quality, increasing its shelf-life by providing safe products. At the same time, the role of the coatings permeable to O_2 and CO_2 is to control the moisture transfer and to reduce surface abrasion [19]. Studies have shown that coated fruits have minimal weight loss [19,20], decreased respiration rate [21], decreased enzyme activity, maintaining visual quality [22], which resulted in a remarkable extension of shelf-life [19,20,22].

Plum is a fruit highly enjoyed by consumers, both fresh and dry, for its characteristic flavor and for its action as a natural laxative. It has high nutritional value, with high sugar, anthocyanins, phenolic compounds concentrations, besides being a source of ascorbic acid and β-carotene [23]. However, plums present a short harvesting period [24] with high perishability due to their fragile structure, high water content and fungal infection, which limit your long-distance transport and storage for longer times. The application of edible coating on the surface of the plums is a viable alternative to maintain or improve their post-harvest quality. Thus, the study of the viability of arrowroot starch application in the development of edible and biodegradable films and coatings, both for the reduction of post-harvest losses of fruits and for the protection of the environment, is essential. The number of research studies that have evaluated the applicability of arrowroot starch in the scientific literature is scarce, making it necessary to carry out new studies that provide information that contribute to its application as a 100% natural edible coating in fruits, such as plums that are highly perishable, as a form of conservation. This evaluation is particularly important, as this starch has great potential for replacing conventional starch due to its functionality as a hydrocolloid, thickening and gelling agent, as well as biodegradable and edible food packaging. In addition, the information found in these studies may be used in future work to design edible starches coatings with improved properties, enabling the expansion of the post-harvest area for other fruits and vegetables. The application of edible fruit coatings has been identified as a low-cost technique and, therefore, feasible for use by small local farmers and small agribusiness, as it uses simple technology and does not require high investment in equipment. Therefore, the aim of this research work was to evaluate the ability to produce edible coatings and films from arrowroot starch. Films were produced with different concentrations of arrowroot starch using glycerol as plasticizer. In addition, their characterization in appearance, thickness, water activity, moisture content,

solubility and vapor permeability were analyzed. Subsequently, arrowroot starch edible coatings were applied to the surface of plums to evaluate their efficiencies as barriers to water loss and consequent reduction the mass loss of fruits during storage at 5 and 25 °C.

2. Materials and Methods

2.1. Materials

In this study, plum fruits (*Prunus domestica*) were purchased from local market of Campinas, São Paulo, Brazil. For the plums coating and preparation of the edible biodegradable films, native arrowroot (*Maranta arundinacea* Linn.) starch was used as film-forming matrix and the glycerol P.A. (Reagen, Quimibrás Indústrias Químicas S.A., Rio de Janeiro, Brazil) as plasticizing agent. The arrowroot starch used in this research work had the same characteristics as the starch used previously in a detailed study of physical-chemical, thermal and micro-structural characterization carried out by our research group [10], as it is the same starch. The arrowroot starch presented 15.24 ± 0.19% of water, 0.40 ± 0.03% of protein, 0.12 ± 0.01% of fat, 0.33 ± 0.01% of ash and 83.91 ± 0.10% of carbohydrates (previously determined according to AOAC [25]), as well as an amylose content of 35.20 ± 1.63% (determined according to the method described by Martínez [26]) [10]. The starch granules presented circular, ellipsoid and oval morphological characteristics of varying sizes; type "C" crystalline structures (determined using the X-ray diffractometer, X'Pert model, Philips Analytical X Ray, Almelo, Netherlands), glass transition temperature (Tg) between 118 and 120 °C (determined using a Differential Scanning Calorimeter, DSC1, Mettler Toledo, Schwerzenbach, Switzerland). The thermogravimetric analysis (performed on a thermogravimetric analyzer, TGA-50 M, Shimadzu, Kyoto, Japan) showed that 40% of the mass loss of starch related to starch depolymerization occurred between 330 and 410 °C, demonstrating that arrowroot starch is thermally stable [10].

2.2. Film Production

The film-forming solution was produced by dispersing the starch in distilled water at concentrations of 1%, 2%, 3%, 4%, and 5% (mass/mass). After complete dispersion, solutions were heated to 85 ± 2 °C in a thermostatic bath (TECNAL, Piracicaba, Brazil) under stirring for approximately 5 min, until complete gelatinization, according to the methodology described by Nogueira et al. [10]. Afterwards, the glycerol was incorporated to the starch solution in a 15% concentration regarding the macromolecule mass and, then, homogenized. Aliquot of 25 mL of the resulting film-forming solution was placed among the support plate (12 cm in diameter) and exposed to drying at 25 ± 5 °C for about 24 h. The dried films were removed from the support plate and conditioned at 25 °C and 55 ± 3% of relative humidity for 48 h, before their characterization.

2.2.1. Visual Aspect

Visual and tactile analyses were made to choose the homogeneous films, with no cracks and flexible for handling.

2.2.2. Film Thickness, Water Activity and Moisture Content

The film thickness was measured using a micrometer (Mitutoyo, model MDC 25M, MFG, Kawasaki, Japan), with an accuracy of ±0.001 mm, in ten different areas of each film. Water activity was measured using an AquaLab Lite water activity meter (Decagon Devices Inc., Pullman, USA), operating at 25 °C, in triplicate. Film samples were exposed to drying in a forced air oven at 105 °C for 24 h, in triplicate, to determine the moisture content [25].

2.2.3. Solubility in Water

The water solubility of the films was quantified following the methodology proposed by Gontard et al. [27]. Dried film samples (105 °C for 24 h) with diameters of 2 cm were individually immersed in 50 mL distilled water and kept under slow mechanical stirring for 24 h at 25 ± 2 °C. Afterwards, the non-solubilized samples were removed, dried (105 °C

for 24 h) and weighted to determine the final dry mass. The water-soluble content was then calculated as the percentage weight that remained after water immersion.

2.2.4. Water Vapor Permeability

The water vapor permeability rate of films was gravimetrically determined, based on ASTM E96-80 method [28]. Samples of each film formulation were fixed individually in acrylic cells, with a central opening (diameter of 4.3 cm) and a bottom filled with dried calcium chloride (0% relative humidity at 25 °C). The cells were placed in a desiccator containing saturated sodium chloride, providing 75% of relative humidity. Water vapor transferred through the film was determined by mass gain of calcium chloride. The cell weight was recorded daily for at least 7 days. The permeability of the film was calculated according to the equation described by Nogueira, Fakhouri and Oliveira [10] using linear regression between weight gain (g) and time (h), to find the slope of the line that determines the amount of water acquired over time.

2.2.5. Microstructure of the Film

The microstructure of the film was observed under a scanning electron microscope (Leo 440i, Electron Microscopy/Oxford, Cambridge, England). Film samples were placed on double-sided carbon adhesive tape adhered to stub, submitted to application of a gold layer (model K450, Sputter Coater EMITECH, Kent, United Kingdom) and observed in scanning electron microscope operated at 20 kV.

2.3. Plums Coating

In the evaluation of plums coating, fruits were sanitized with chlorinated water and dried. Coatings were prepared in aqueous solution in the following concentrations: control—distilled water; 2% arrowroot starch (mass/mass) plus 15% glycerol (mass/starch mass); 4% arrowroot starch (mass/mass) plus 15% glycerol (mass in relation to starch mass). The coatings were prepared by dispersing the starch in distilled water. After complete dispersion, the solutions were heated to 85 ± 2 °C in a thermostatic bath (TECNAL, Piracicaba, Brazil), in constant agitation for 5 min, until complete gelatinization [10]. Then, glycerol was added to the starch solution and homogenized. The fruit were completely immersed in the respective solutions for 1 min and then placed on metal wire screens with ventilation incidence, for a period of 12 h, in order to dry the coating. Afterwards, they were packed in polyethylene lid trays, homogeneously with five fruits per pack and stored at 5 and 25 °C for 35 days.

2.3.1. Appearance and Mass Loss

Plums were selected for the absence of physiological defects, fungal deterioration, holes and rot. In the mass loss analysis, three replications were used in each treatment, i.e., three packs containing five plums each. Plums were weighed every 7 days during the storage period to determine weight loss. Results were expressed as the percentage of weight loss based on the initial weight.

2.3.2. Respiratory Rate

Fruits were packed in glass containers with capacity of 600 mL and hermetically sealed for 30 min. The respiratory rate was determined by CO_2 production, in duplicate. The gas from the free space of the glass container, used in the packaging of samples, was circulated through Agri-Datalog's O_2 and CO_2 electronic analyzers. Based on CO_2 concentration, free space volume, fruit mass and closing time, respiration was calculated, with values expressed in milligrams of CO_2 kg^{-1} h^{-1}.

2.3.3. pH and Soluble Solids

The pH was measured by direct reading of the homogenized fruit utilizing a potentiometer (Digimed pH meter DM-20) and the soluble solids using an Abbé type bench refractometer, following AOAC's official methodology [29], in triplicate.

2.3.4. Moisture Content

The determination of the moisture content of samples was carried out by drying them in an air-forced oven at 105 °C for 24 h [29], in triplicate.

2.3.5. Titratable Total Acidity

The total titrable acidity of the plums was determined in triplicate, by the titration of 10 g of crushed pulp and homogenized with 90 mL of distilled water using standard sodium hydroxide solution (NaOH 0.1 N) titrator, to end point of pH 8.1 using the potentiometer (Digimed pH meter DM-20) as a turning point indicator and the results expressed as a percentage of citric acid.

2.3.6. Anthocyanins Content

The determination of the anthocyanins content of plums was carried out following the methodology described by Sims and Gamon [30], with adaptations. The plum samples were homogenized with 3 mL of cold acetone/Tris-HCl solution (80:20, volume/volume, pH 7.8 0.2 M) for 1 min. After 1 h at repose in the absence of light, the samples were centrifuged for 15 min at 3500 rpm and the supernatants read on the spectrophotometer (model B422, Micronal) in the visible region at 537 nm (anthocyanins). The acetone/Tris-HCl solution was used as blank sample. Absorbance values were converted to mg g^{-1} of fresh sample.

2.4. Statistical Analysis

The SAS program was used for calculating analysis of variance (ANOVA) and the Tukey test to evaluate differences between means in the 95% confidence interval.

3. Results and Discussions

3.1. Characterization of Films

After drying, all films, except those with 1% concentration of the arrowroot starch, could be removed from the support plates without tearing, being able to manipulation without risk of ruptures or areas prone to cracking. All films were homogeneous, without bubbles and insoluble particles visible to the naked eye.

The starch concentrations used in the formulation did not affect the visual appearance of the films. All the films were transparent, odorless and good-looking, similar to petroleum-based plastic films, as can be seen in Figure 1. Arrowroot starch films showed one bright face (surface contact with the plate dish during the drying period) and the other one was matte (surface exposed to air during drying). The same was reported by Basiak et al. [31] for wheat, corn and potato starch films.

The films thickness increased significantly ($p < 0.05$) from 0.029 ± 0.01 to 0.101 ± 0.14 mm, with the concentration increasing from 1 to 5% of arrowroot starch in solution, as shown in Table 1. Films with thicknesses of 0.062 ± 0.008 mm were produced with 2 wt % of the arrowroot starch and 17 wt % of the glycerol by Oliveira Filho et al. [32]. The thicknesses of arrowroot starch films produced by Nogueira, Fakhouri and Oliveira [10] ranged from 0.026 ± 0.008 to 0.082 ± 0.011 mm, when they used starch concentrations ranging from 2.59% to 5.41% (mass/mass) and concentrations of glycerol from 9.95% to 24.08% (mass/starch mass). The increase in thickness is due to an increase in the amount of dry matter, in the same volume of film-forming solution, deposited per unit area per support plate. According to Sobral [33], obtaining films by casting method generates films of different thicknesses depending on the mass applied on the support, shape of the support and its flat even surface (local thickness variations can be caused by unevenness).

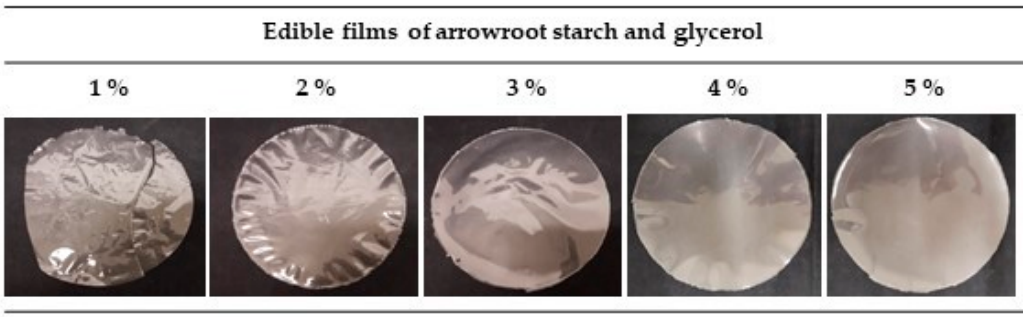


Figure 1. Images of the films with different concentrations of arrowroot starch in film-forming solution.

Table 1. Thickness, water activity (Aw), moisture content, water in solubility and water vapor permeability values obtained for films produced with different concentrations of arrowroot starch in film-forming solution.

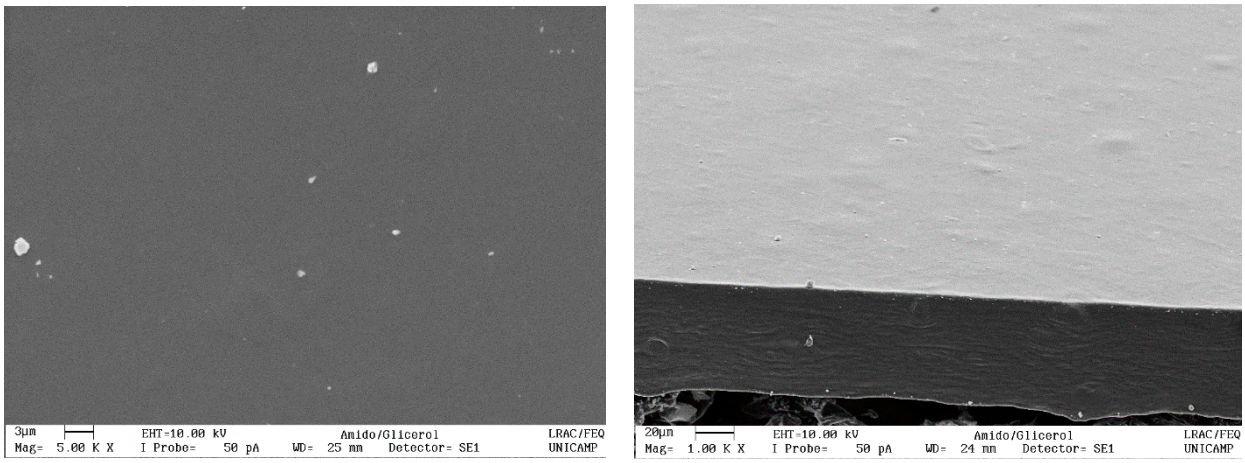
Films	Thickness (mm)	A_W (decimal)	Moisture Content (%)	Solubility in Water (%)	Water Vapor Permeability (g mm/m^2 day kPa)
1%	0.029 ± 0.01 b *	0.594 ± 0.024 a	3.22 ± 0.25 b	22.45 ± 1.21 a	2.20 ± 0.06 bc
2%	0.059 ± 0.01 ab	0.500 ± 0.008 cb	6.44 ± 0.83 ab	15.30 ± 1.74 b	1.70 ± 1.15 c
3%	0.053 ± 0.01 ab	0.452 ± 0.016 c	7.27 ± 0.30 a	14.19 ± 0.77 b	1.97 ± 0.44 bc
4%	0.081 ± 0.01 a	0.457 ± 0.026 cb	5.42 ± 1.53 ab	16.11 ± 0.73 b	4.08 ± 0.63 a
5%	0.101 ± 0.14 a	0.511 ± 0.028 b	7.95 ± 2.19 a	13.89 ± 0.65 b	3.68 ± 0.17 ba

* Means and standard deviation followed by the same lowercase letter in column did not differ statistically from each other by the Tukey test at $p > 0.05$.

Table 1 shows arrowroot films water activity and moisture content values. The water activity presented by the films ranged from 0.45 to 0.59. It is important that edible films have low water activity values to decreased microbiological proliferation at room temperature and usual conditions of relative humidity [34]. As the films presented water activity values below of the 0.60 (Table 1), it can be considered that they are microbiologically stable, as there is no microbial growth below this value, ensuring greater food security so that it is consumed.

The films presented water contents ranging from 3% to 8%, lower than reported by Oliveira Filho et al. [32] of 14.2 ± 0.9% for arrowroot starch films (2 wt % of the starch and 17 wt % of the glycerol). Colussi et al. [35] obtained water contents ranging from 13.49% to 23.42% for native and acetylated rice starch films with medium and high amylose contents, conditioned at 65% of relative humidity and at 21 °C. According to Sarantópoulos et al. [36], the film must have maximum water content of 10% in order to have good barrier properties.

Films with starch concentrations higher than 1% presented statistically significant lower values ($p < 0.05$) for solubility in water, compared to 1% films (Table 1). These films were less soluble than arrowroot starch and cassava starch films which presented solubility of 60.7% [32] and 27.5% [21], respectively. The decrease in the water solubility of the film is associated with a formation of a more compacted structure, indicating a greater number of bonds between chains, i.e., a more resistant three-dimensional matrix. The increase of the starch concentration in the solution was able to produce films with structured, organized and compacted chains, which probably made it difficult the access of water molecules to the hydrophilic groups due to least mobility, reducing solubility [37]. This can be confirmed in Figure 2a,b by SEM images of the surface and cross-section of films. The film presented a smooth and homogeneous surface, with a continuous and dense network formed by arrowroot starch, which resulted in better water barrier properties. This property directs the application of the film as food packaging. In some cases, their total solubilization in water may be beneficial, as in semi-ready products intended for preparation under cooking, or when it will be consumed with the product. For applications such as edible coating, it is desirable that coating is completely dissolved when consumed with food. However, when the food is liquid or exudes an aqueous solution, high solubility films are not indicated [3].



(**a**) (**b**)

Figure 2. Scanning electron microscopy (SEM) images: (**a**) surface and (**b**) cross section of the film produced with 4% (mass/mass) of arrowroot starch and 15% of glycerol (mass/mass of starch).

The water vapor permeability of films varied from 1.70 ± 1.15 g mm/m^2 day kPa (2% starch film) to 4.08 ± 0.63 g mm/m^2 day kPa (4% starch film). Films of native and acetylated rice starch with medium and high amylose contents showed water vapor permeability ranging from 5.33 to 10.33 g mm/m^2 day kPa [35]. Water vapor permeability is the measure of the amount of moisture that passes through the unit area of material per unit time [38]. This moisture transfer usually occurs through the hydrophilic portion of a barrier [39]. The natural polymers used for making edible films are generally hydrophilic, such as starch. They contain polar groups that interact with permeant water molecules inducing plasticization during permeation. The sorption of water that occurs during the permeation process increases the polymer free volume allowing the polymer chain segments to increase mobility due to swelling. The higher the mobility, the higher the water vapor permeability [38]. Thus, an increase in the water vapor permeability of the films can be observed with increasing starch concentration from 1% to 5% and its thickness (Table 1). Park and Chinnan [40] observed that the water vapor permeability increased with the thickness due to a linear behavior between this property and the thickness of the films. It can be seen that the 5% starch film showed a reduction in water vapor permeability compared to the 4% film, despite being significant. This reduction is a consequence of the variability in the thickness of the 5% films.

The increase in water vapor permeability with starch concentration has been reported for films made from native and dual-modified yam (*Dioscorea rotundata)* starch films [41]. Whereas water vapor permeability is the result of sorption and moisture diffusivity in solid matter, starch is likely to increase the affinity of the film and water or induce a decrease in local viscosity and then increase diffusivity [38]. Incorporation of microemulsions and nano-emulsions of carnauba wax in arrowroot starch films significantly reduced their permeability to water vapor and water solubility, due to the hydrophobic feature of the wax [32].

3.2. Plums Coating

The concentrations of 2% and 4% of arrowroot starch in film-forming solution were chosen to be tested as edible coating on plums, in order to evaluate their efficiencies as barriers to water loss and, consequently, reducing the mass loss of fruits during storage at 5 and 25 °C. The 1% starch concentration has not been tested, as it produces a very liquid coating solution, which could hinder its adherence evenly on the fruit surface. On the other hand, the 5% arrowroot starch concentration produced a very viscous coating solution, which could generate a very thick and opaque coating on the fruit surface, which could alter its appearance and impair its acceptance by the consumer.

Arrowroot starch coatings adhered well to the peels of the plums. However, the 4% film was opaque and flocculated, principally after storage at 25 °C (Figure 3). Opacity may vary depending on the amylose content of starches, as their solution molecules, due to their linearity, tend to orientate parallel enough to form hydrogen bridges between adjacent chain hydroxyls. As result, the affinity of the polymer for water is reduced, favoring the formation of opaque pastes and resistant films [3,42]. In addition, it is possible that during the storage of plums at room temperature, the coating has lost water to the environment favoring flocculation.

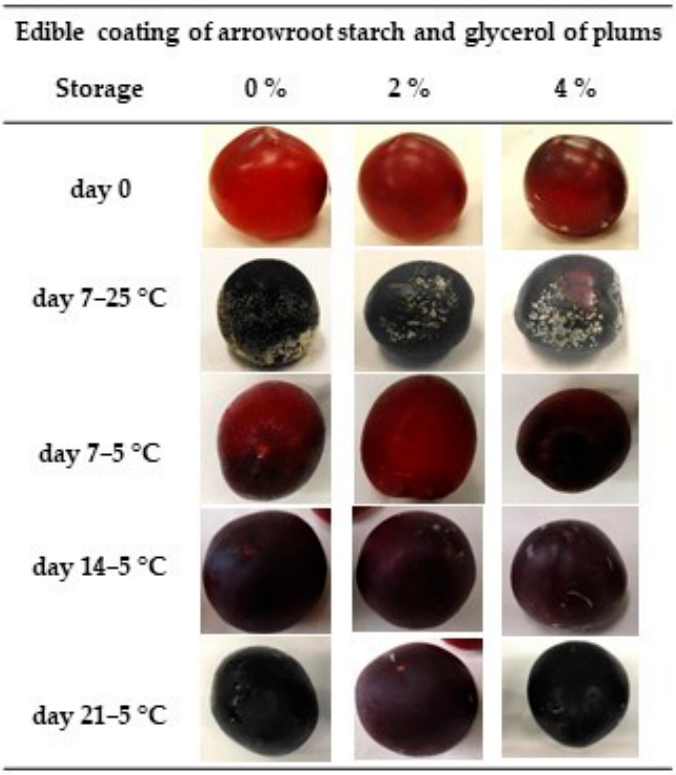


Figure 3. Images of plums with different concentrations (0, 2 and 4% mass/mass) of edible arrowroot starch coating stored at 25 °C and 5 °C.

The plums coated with the 2% starch concentration were brighter. No visible changes in the texture of the plums were observed when stored at 5 °C, unlike those maintained at 25 °C which, after 7 days of storage, were wilted and showed growth of microorganisms visible to the naked eye.

Regardless of treatment, plums stored at 5 °C for 14 days still showed firm consistency, with no incidence of surface cracks, growth of microorganisms visible to the naked eye or presence of liquid in the packages. Only after 21 days of storage, for all treatments, it was possible to observe the fungal growth and rot incidence in just some samples, although most of them still presented firm consistency and absence of microbiological contamination. The shelf-life of plums stored at 5 °C was 35 days, while for the plums stored at a temperature of 25 °C, it was only 7 days.

The incorporation of antimicrobial agents into the starch coatings could avoid the microbiological contamination of the fruit, prolonging its shelf-life. Edible films based on pea starch and guar gum incorporated with epigallocatechin-3-gallate and two native Australian plants—blueberry ash fruit and macadamia—showed antimicrobial properties and could be used to preserve food safety and prolong the shelf-life of packaged products [43].

During the storage period, there was a significant loss of mass of the evaluated plums, and the highest values were observed in fruit kept at 25 °C, after 7 days of storage, compared with fruit stored at 5 °C (Table 2). The 4% starch coating significantly reduced

the mass loss of plums stored at 25 °C, compared to the others. There was a significant mass loss decrease of the plums with reduced storage temperature. Among the studied coatings, the largest mass loss was observed for uncoated plums (10.63 ± 0.27%), followed by plums coated with 4% (8.37 ± 1.29%) and 2% (7.18 ± 1.53%) starch concentration in film-forming solution, stored at 5 °C after 35 days. The edible starch coating was effective in reducing plum weight loss over time, when compared to uncoated plums (Table 2). Garcia et al. [44] also observed a reduction over time in the weight loss of strawberries that received coatings containing 2% of corn starch. This can be explained by the increased difficulty for water migration from the fruit to the environment, caused by the film formed around the fruit. Films with 2% of arrowroot starch produced the lowest water vapor permeability rate. On the other hand, the flocculation of coating with 4% starch promoted discontinuity in the structure of the film formed on plum surface, which resulted in an impaired barrier property, in the long term. Therefore, it can be concluded that the coating of plums with film-forming solution in the concentration of 2% of arrowroot starch in solution acted as a barrier to water loss, consequently reducing the loss of mass of the fruit during its storage, mainly when associated with storage under refrigeration.

Table 2. Mass loss (%) of plums with different concentrations of edible coating, stored for up to 7 days at 25 °C and 35 days at 5 °C.

Treatments	Storage (Days)					
	0	7	14	21	28	35
0%—25 °C	0.00 ± 0.00 aB *	5.43 ± 0.34 aA	-	-	-	-
2%—25 °C	0.00 ± 0.00 aB	5.40 ± 0.85 aA	-	-	-	-
4%—25 °C	0.00 ± 0.00 aB	2.77 ± 0.31 bA	-	-	-	-
0%—5 °C	0.00 ± 0.00 aD	0.00 ± 0.08 cD	1.74 ± 0.23 aC	2.62 ± 0.31 aC	8.50 ± 0.82 aB	10.63 ± 0.27 aA
2%—5 °C	0.00 ± 0.00 aC	0.02 ± 0.03 cC	2.38 ± 0.17 aBC	3.57 ± 0.11 aB	4.90 ± 0.30 aBA	7.18 ± 1.53 aA
4%—5 °C	0.00 ± 0.00 aB	0.55 ± 0.59 cB	2.37 ± 0.32 aB	3.33 ± 0.51 aB	7.78 ± 2.51 aA	8.37 ± 1.29 aA

* Averages followed by the same lowercase letter in column and uppercase in line do not differ statistically from each other to the Tukey test at $p > 0.05$.

The respiration rate of plums stored at 25 °C and 5 °C is shown in Table 3. The lowest values of respiratory rates were observed in plums stored at 5 °C, regardless of treatment. For all treatments stored at 5 °C, a significant reduction in respiration rate was observed on 14 days of the experiment. After 35 days at 5 °C, the fruit showed a significant decrease of respiratory rate in relation to day 0. The respiration rate of vegetables is affected by storage temperature and microbiological contamination, as well as the action of coating. The reduction of the storage temperature reduces the cellular metabolism [45] and consequently the respiratory rate of the fruit [24,46]. Regarding treatments, no significant influences ($p > 0.05$) were observed in respiratory rate. These results indicate that arrowroot starch coatings had a semipermeable characteristic, because fruit continued to breathe, perspiring and losing water, but in smaller quantity when stored at low temperature.

Table 3. Respiratory rate (mg CO_2 kg^{-1} h^{-1}) of plums with different concentrations of edible coating stored for up to 7 days at 25 °C and 35 days at 5 °C.

Treatments	Storage (Days)					
	0	7	14	21	28	35
0%—25 °C	247.84 ± 14.02 aA *	252.3 ±5.09 aA	-	-	-	-
2%—25 °C	247.84 ± 14.02 aA	235.60 ± 48.05 bacA	-	-	-	-
4%—25 °C	247.84 ± 14.02 aA	239.18 ± 26.95 baA	-	-	-	-
0%—5 °C	247.84 ± 14.02 aA	122.25 ± 4.75 cC	148.11 ± 2.99 bCB	152.96 ± 7.30 bCB	178.32 ± 8.62 aB	145.17 ± 9.83 aCB
2%—5 °C	247.84 ± 14.02 aA	223.53 ± 25.20 bacBA	170.90 ± 4.40 baBC	184.28 ± 0.20 baBC	187.29 ± 3.24 aBC	157.83 ± 10.42 aC
4%—5 °C	247.84 ± 14.02 aA	127.37 ± 6.27 bcD	180.08 ± 6.59 aCB	207.47 ± 6.49 aB	175.24 ± 12.53 aCB	166.84 ± 1.13 aC

* Averages followed by the same lowercase letter in column and uppercase in line do not differ statistically from each other to the Tukey test at $p > 0.05$.

In Figure 4a,b, a significant ($p < 0.05$) increase in pH and soluble solids and a decrease in titratable total acidity of the plum pulp can be observed at the end of the experiment regarding to day 0, for all treatments regardless of storage temperature. Valero et al. [47], evaluating the effects on four plum cultivars coated with alginate (1% and 3%), for 35 days at 2 °C, reported variations of acidity between cultivars with delayed acidity decrease in the treatment coated with 1% and 3% of alginate, although they did not observed significant differences between these treatments.

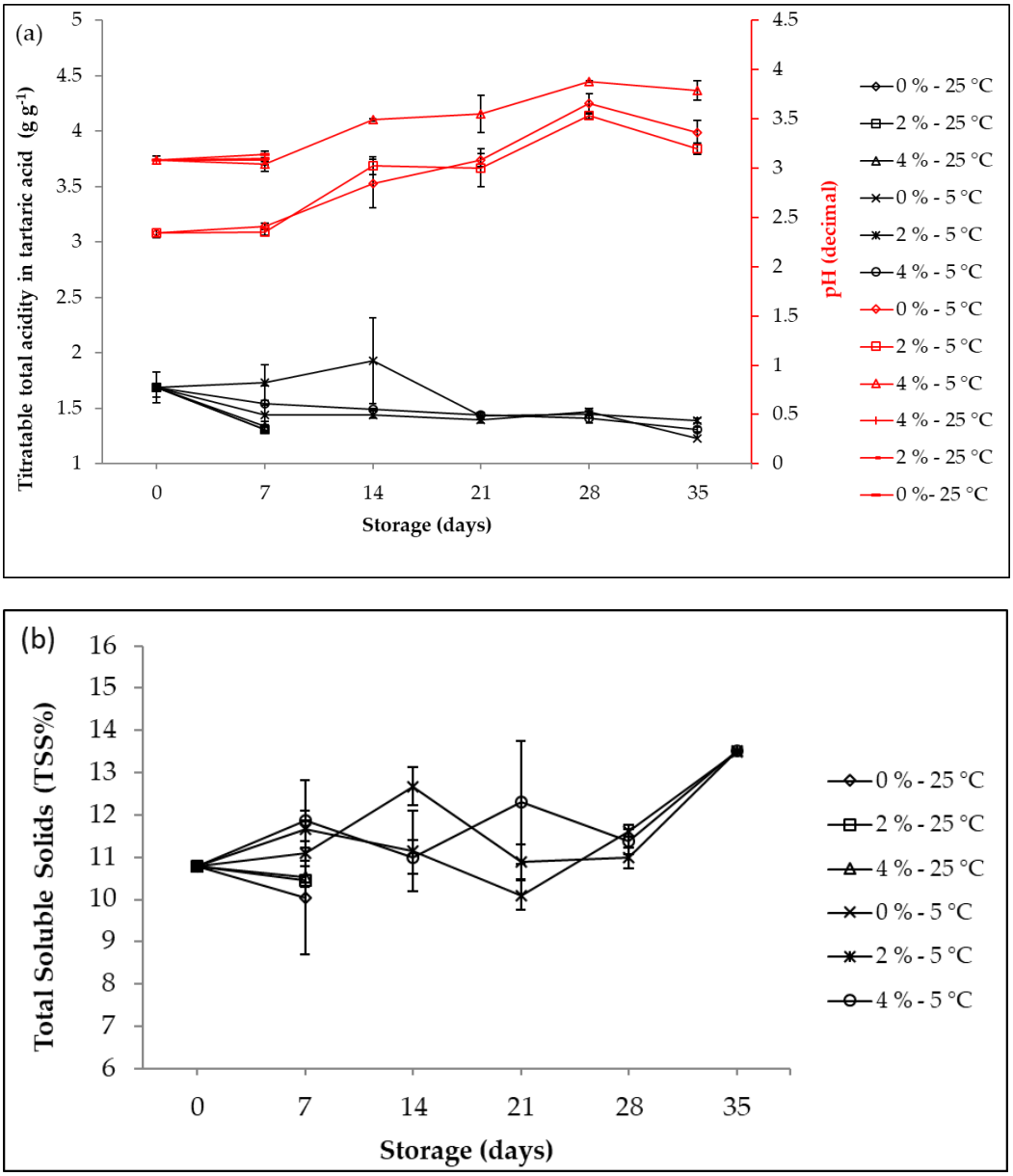


Figure 4. Titratable total acidity in tartaric acid (g g^{-1}) and pH (decimal) (**a**) and total soluble solids (TSS%) (**b**) of plums with different concentrations of edible coating stored for up to 7 days at 25 °C and 35 days at 5 °C.

In this work, the plum coating did not significantly influence these parameters. The increase of pH and soluble solids contents and decrease of acidity may be associated with the fruit ripening process, with degradation of the starch into glucose by glycolysis. According to Chitarra and Chitarra [48], soluble solids content increases as fruit ripens due to increased synthesis or degradation of polysaccharides and accumulation of sugars. The

soluble solids present in fruit are important compounds (mainly sugars and organic acids) responsible for the taste and consequent acceptance of the product by consumers.

A high-water content was found in the pulp of plums, with variation of 83.27 ± 4.55% to 89.32 ± 0.05%. Concerning anthocyanins content, there was a statistically significant increase ($p < 0.05$) over plum storage days, at 25 °C and 5 °C in relation to day 0, regardless of treatment (Table 4). This happens because, during the fruit ripening process, there is a reduction of the chlorophyll pigment and an increase of anthocyanins, which is evidenced by change in the color of the fruit from green to red [48]. The higher anthocyanins content found on day 14 for plum samples stored at 25 °C may be explained by the higher respiration rate presented by these samples. Higher respiration rate results in higher metabolism, which can lead to higher pigment production [48]. Cordenunsi et al. [49] found a significant increase ($p < 0.05$) in the anthocyanins content of cv Oso Grande strawberries, harvested with 75% red surface color and stored at 16 °C and 25 °C.

Table 4. Anthocyanins content (mg g^{-1} of fresh pulp) of plums with different edible coating concentrations, stored for up to 7 days at 25 °C and 35 days at 5 °C.

Treatments	Storage (Days)					
	0	7	14	21	28	35
0%—25 °C	2.45 ± 0.16 aB *	3.47 ± 0.25 aA	-	-	-	-
2%—25 °C	2.45 ± 0.16 aB	3.67 ± 0.37 aA	-	-	-	-
4%—25 °C	2.45 ± 0.16 aB	3.08 ± 0.28 aA	-	-	-	-
0%—5 °C	2.45 ± 0.16 aA	3.12 ± 0.04 aA	2.15 ± 0.24 aA	2.00 ± 0.16 aA	2.65 ± 0.79 bA	2.88 ± 0.29 aA
2%—5 °C	2.45 ± 0.16 aC	3.19 ± 0.21 aBAC	2.69 ± 0.24 aBC	2.88 ± 0.27 aBC	3.42 ± 0.08 baBA	3.87 ± 0.62 aA
4%—5 °C	2.45 ± 0.16 aC	3.17 ± 0.16 aBAC	2.15 ± 0.37 aC	2.55 ± 1.07 aBC	3.85 ± 0.44 aBA	3.96 ± 0.10 aA

* Averages followed by the same lowercase letter in column and uppercase in line do not differ statistically from each other to the Tukey test at $p > 0.05$.

4. Conclusions

Edible films with increasing concentrations from 1% to 5% of arrowroot starch in solution were produced by a casting method with good handling characteristics, transparency and colorlessness. Films produced with 1% starch tore when removed from the support plate and were fragile to handle. An increasing concentration of starch in the solution increased the thickness of the films and decreased their water solubility property. On the other hand, it impaired the water vapor barrier properties of the films.

The application of the edible coating of arrowroot starch to plums was effective in reducing the mass loss of fruits, principally when 2% of starch concentration was used associated with storage temperature of 5 °C. The 2% starch coating adhered well to the surface of plums and made plums brighter, whereas the 4% starch coating appeared opaque and flocculated. Furthermore, arrowroot starch coatings did not present significant effects on the physicochemical parameters of the fruit such as respiratory rate, pH, total titratable acidity, soluble solids and anthocyanin content, which were more influenced by the storage temperature. The plums stored at 25 °C withered and showed fungal growths that were visible to the naked eye after 7 days. For fruits stored at 5 °C, no visible changes in texture were observed. The lowest values of mass loss and respiratory rate in plums were obtained when stored at 5 °C and for plum samples with 2% starch edible coatings. There was an increase in pH, soluble solids content and anthocyanins, and a decrease in the acidity of the plums after storage, due to the fruit ripening process. It could be concluded that the use of coating allied to the low temperature favors the reduction of the mass loss of plums during storage.

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review and editing, G.F.N., C.T.S., F.M.F. and R.A.d.O.; visualization, G.F.N., C.T.S., F.M.F. and R.A.d.O.; supervision, G.F.N. and R.A.d.O.; project administration, G.F.N. and R.A.d.O.; funding acquisition, G.F.N. and R.A.d.O. All authors have read and agreed to the published version of the manuscript.

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