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Development and optimization of biodegradable films based on achira flour

Margarita María Andrade-Mahecha[a], Delia Rita Tapia-Blácido[b], Florencia Cecilia Menegalli[a,*]

[a] Food Engineering Departament, State University of Campinas, UNICAMP, Rua Monteiro Lobato n° 80, Barão Geraldo, 13083-862 Campinas, SP, Brazil
[b] Departamento de Química, Faculdade de Filosofia, Ciências e Letras, Universidade de São Paulo, Av. Bandeirantes, 3.900, 14040-901 Ribeirão Preto, SP, Brazil

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ABSTRACT

The influence of glycerol concentration (C_g), process temperature (T_p), drying temperature (T_s), and relative humidity (RH) on the properties of achira flour films was initially assessed. The optimized process conditions were C_g of 17 g glycerol/100 g flour, T_p of 90 °C, T_s of 44.8 °C, and RH of 36.4%. The films produced under these conditions displayed high mechanical strength (7.0 MPa), low solubility (38.3%), and satisfactory elongation values (14.6%). This study showed that achira flour is a promising source for the development of biodegradable films with good mechanical properties, low water vapor permeability, and solubility compared to films based on other tubers.

1. Introduction

Concentrated efforts toward the development of biodegradable materials with properties that ensure food safety and security while minimizing the environmental impact of their use has been the focus of different studies. To produce such films, researchers have usually employed commercial biopolymers and lipids, which are then mixed during film processing (Arvanitoyannis, 1999; Arvanitoyannis & Biliaderis, 1998, 1999; Arvanitoyannis, Nakayama, & Aiba, 1998; Arvanitoyannis, Psomiadou, & Nakayama, 1996; Psomiadou, Arvanitoyannis, & Yamamoto, 1996). Results have not always been favorable due to the thermodynamic incompatibility of biopolymers, which may also cause phase separation (Grinberg & Tolstoguzov, 1997). To overcome this problem, researchers have turned to natural mixtures of starch, protein, lipids, and fibers, which can be obtained in the form of flour from raw materials of plant origin such as cereals and legumes. The use of natural mixtures (flours) from agricultural crops has also attracted increasing attention because of the several botanical sources available as material for the development of edible films, allied with the possibility of improving their mechanical and barrier properties. The characteristics of the films based on flour are a result of the natural interactions occurring between the starch, protein, and lipids during drying of the casting suspension (Tapia-Blácido, Mauri, Menegalli, Sobral, & Añón, 2007), which can also be influenced by the different variables employed during the production process (Araujo-Farro, Podadera, Sobral, & Menegalli, 2010; Dias, Müller, Larotonda, & Laurindo, 2010; Gontard, Guilbert, & Cuq, 1993; Parris, Dickey, Kurantz, Moten, & Craig, 1997; Tapia-Blácido, Sobral, & Menegalli, 2005a, 2011). The functional properties of the films can also be modified by changing the temperature and relative humidity applied during the drying step (Alcantara et al., 1998; Arvanitoyannis et al., 1998, 1996; Denavi et al., 2009; Tapia-Blácido, Sobral, & Menegalli, 2005b, among others).

Tubers and roots are important sources of flours in tropical regions of Latin America, the Caribbean, Africa, and Asia. Among these sources, achira (*Canna indica* L.) is a widely cultivated plant in different countries of Latin America and Asia, and it has been commercially employed for starch extraction (Leonel, Sarmento, Cereda, & Guerreiro, 2002; Peroni, Rocha, & Franco, 2006). Several studies have reported on some interesting features of the achira starch, such as paste clarity, high viscosity, and high resistance to hydrolysis by α-amylase (Hung & Morita, 2005; Puncha-Arnon, Puttanlek, Rungsardthong, Pathipanawat, & Uttapap, 2007; Santacruz, Ruales, & Eliasson, 2003). The high viscosity and clear paste of the achira starch makes it a potentially useful thickening agent (Puncha-Arnon et al., 2007) and may lead to its application in the production of excellent transparent noodle (Hung & Morita, 2005). Other studies have indicated that the achira starch has high amylose content and high level of phosphorus, as compared to starch obtained from other commercially important tubers, like potato and cassava. These components play an important role in the functional properties of the achira starch (Cisneros, Zevillanos,

* Corresponding author. Tel.: +55 19 3521 4039, fax: +55 19 3521 4027.
E-mail address: fcm@fea.unicamp.br (F.C. Menegalli).

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& Cisneros-Zevallos, 2009; Peroni et al., 2006; Piyachomkwan et al., 2002; Thitipraphunkul, Uttapap, Piyachomkwan, & Takeda, 2003). Considering these properties, it is surprising that no studies on the production of films from achira flour have been carried out to date. Only the starch obtained from this tuber has been explored in an *in vitro* investigation on the digestibility of edible films (Hernández, Emaldi, & Tovar, 2008). The present study aims to determine the optimal formulation of achira flour film by using response surface methodology and multi-response analysis, in order to obtain films with low solubility, moderate elongation, and high resistance to break.

2. Material and methods

2.1. Raw material

Achira flour was obtained from rhizomes cultivated in humid subtropical climate (Köppen *Cwa*) in the city of Conchal, SP, Brazil (22°19′48″S, 47°10′22″W, 591 m.a.s.l.). Seven-month-old rhizomes were harvested and supplied by Corn Products Brazil. The rhizomes were cleaned and peeled manually, cut into 3-mm slices, and immersed into $K_2S_2O_5$ solution (0.8%, w/v), to prevent oxidation of the slices. Then, the slices were dried at 40 °C for 45 h in a temperature-controlled oven (model MA 415UR, Marconi, Piracicaba, Brazil). Finally, dry-milling and sieving (115/400 mesh) were used to obtain the achira flour with granulometry ≤43.5 μm.

2.2. Chemical analysis

The moisture, crude protein, total dietary fiber, and ash contents of the achira flour were determined using AOAC methods (2005); the starch content was calculated by the method of Diemair (1963). The Bligh and Dyer (1959) method was used for determination of the total lipid content. The amylose content was obtained by following the ISO 6647 method and is expressed as the percentage (%) of total dry starch content. All the analyses were performed in triplicate.

2.3. Preparation and conditioning of the flour films

The films were prepared according to the casting technique. To this end, an aqueous suspension of achira flour (4% w/w) was magnetically stirred for 30 min, at room temperature, and then heated up to the processing temperature (T_p: 70, 75, 80, 85, or 90 °C), while gentle stirring was maintained, in order to avoid the formation of bubbles. When the solution reached the T_p value, glycerol was added as a plasticizer (C_g: 15, 20, 25, 30, or 35 g glycerol/100 g flour). After this the film-forming suspension was kept at the processing temperature for 15 min. Next, 76 ± 0.5 g of the suspension was poured and spread on Teflon plates (18 × 21 cm) and dried at 35, 40, 45, 50, or 55 °C and 30, 40, 50, 60, or 70% RH in an oven (Model MA-415UR, Marconi, Brazil) equipped with a control system for drying temperature (T_s) and relative humidity (RH). Prior to the characterization, the dried films were peeled from the Teflon plates, cut into specific shapes for various tests, and stored at 25 °C and 58% RH for 48 h in desiccators containing a saturated NaBr solution. Film thickness was measured with the aid of a micrometer (model FOW72-229-001, Fowler, Newcastle, CA) and was determined as a random average of 15 measurements made at five different regions of the film.

The values of glycerol content (C_g), process temperature (T_p), drying temperature (T_s), and relative humidity (RH) varied according to four variables-five levels of a Central Composite Design (CCD), as reported in Table 1. Preliminary tests were performed under process conditions at the CCD central point, to determine the ideal moisture content of the films. For this purpose, a film sample was weighed every 15 min, until a condition in which the film was easily removed from the teflon plates without any damage was reached.

2.4. Mechanical properties

The mechanical tests were conducted on a texture analyzer TA.XT2i (SMS, Surrey, England). The tensile strength and elongation at break were obtained according to the ASTM D882-02 method (ASTM, 2002). The secant modulus is the slope of the line connecting the origin and a given point on the stress–strain curve, and it was calculated by dividing the corresponding stress value by the designated strain (1%) on the stress–strain curve. This calculation was performed using the software Exponent (SMS, 2003). Five measurements were accomplished for each mechanical test.

2.5. Solubility in water and moisture content

The solubility in water was calculated as the percentage of dry matter of the solubilized film after immersion for 24 h in water at 25 ± 2 °C (Gontard, Guilbert, & Cuq, 1992). Film discs (diameter = 2 cm) were cut, weighed, immersed into 50 mL distilled water, and slowly and periodically agitated. The amount of dry matter in the initial and final samples was determined by drying the samples at 105 °C for 24 h.

The moisture content of the film was measured in samples (2 g) collected at the end of the drying process and after conditioning, by using the ASTM D644-99 methodology (ASTM, 1999). Three measurements were accomplished for each determination.

2.6. Color and opacity

The color, represented as color difference (ΔE^*), was determined according to Gennadios, Weller, Handa, and Froning (1996), and the opacity was analyzed by means of the HunterLab method (Sobral, 1999). A colorimeter (HunterLab, model Miniscan XE) was employed in both cases. The difference in color was calculated as:

$$\Delta E* = \sqrt{(\Delta L*)^2 + (\Delta a*)^2 + (\Delta b*)^2}, \quad (1)$$

where ΔL^*, Δa^*, and Δb^* are the differentials between the color parameter of the samples and of the white standard ($L^* = 93.49$, $a^* = -0.77$, $b^* = 1.40$) used as film background.

2.7. Scanning electron microscopy (SEM)

SEM analyses were carried out as described by Tapia-Blácido et al. (2007). Film samples were maintained in a desiccator with silica gel for 7 d. Film pieces (4 mm × 4 mm) were then mounted on cylindrical aluminum stubs using a double-sided cupper tape, and coated with gold in a VG Microtech (Cambridge, UK) model SC 7620 sputter coater. The film specimens were finally observed under a JEOL Model JSM-5800LV scanning electron microscope, at an accelerated voltage of 10 kV.

2.8. Water vapor permeability

The water vapor permeability (WVP) test was performed using a modified E96-95 ASTM Standard method (ASTM, 1995) at 25 ± 2 °C. Film samples were sealed over the circular opening of a permeation cell containing silica gel, and the cells were then placed in desiccators containing distilled water. The weight loss of the cells was monitored every 1 h, for 9 h.