



The effect of fatty acids on the physicochemical properties of edible films composed of gelatin and gluten proteins



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ABSTRACT

In this study, edible films were prepared with different wheat gluten (GLU) and gelatin (GEL) ratios by the solvent casting technique using glycerol (GLY) or sorbitol (SOR) as plasticizers. Fatty acids (caproic, caprylic, capric, lauric, myristic or palmitic) were also added to a previously selected GLU/GEL/sorbitol film, and the effect of GLU:GEL proportion and type of fatty acid on the film properties were tested. Films plasticized with GLY presented a more significant reduction in the elongation at break (EB). In contrast, films plasticized with SOR did not show a significant difference in the EB. The film's water vapor permeability (WVP) and acid solubility increased with increasing proportions of GEL while the water solubility was decreased. On the other hand, the elongation at break of the films decreased with increasing GEL content, which may be associated with its more rigid structure. The addition of fatty acids resulted in lower WVP and the plasticizing effect was dependent on the degree of interaction with the proteins of the film (identified by thermal analysis). The GLU:GEL proportion and the type of fatty acid affect the film properties (mechanical, solubility, opacity, water vapor barrier), allowing the development of new materials with different and useful functional properties according to the desired application.

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1. Introduction

Edible packaging has been recognized as a potential alternative to conventional packaging since it reduces waste and may improve product stability, quality and safety (Janjarasskul & Krochta, 2010). They provide a semi-permeable barrier against oxygen, carbon dioxide and moisture, reducing respiration, water loss and oxidation rates (Lee, Park, Lee, & Choi, 2003). These last generations of edible films are not expected to replace petrochemical polymer films, but they can help in limiting moisture, aroma, and lipid migrations within composite foods or between food and headspace of conventional packaging (Benbettaieb, Gay, Karbowiak, & Debeaufort, 2016; Vieira, Silva, Santos, & Beppu, 2011). Edible coatings may be

prepared from proteins (eg. gluten and gelatin), polysaccharides, lipids and combination of edible constituents (Fakhoury et al., 2012; Han, Hwang, Min, & Krochta, 2008; Martelli et al., 2017; Moore, Martelli, Gandolfo, Sobral, & Borges Laurindo, 2006).

Gluten proteins are plant proteins and consists of monomeric gliadins and polymeric glutenins (Shewry, Halford, Belton, & Tatham, 2002). They are divided into two groups based on their extractability in aqueous alcohols: the gliadins (extractable) and glutenins (no extractable) (Shewry & Tatham, 1997). The cohesiveness and elasticity of gluten provide integrity and facilitate film formation. Wheat gluten-based films exhibit significant gas-barrier properties and selectivity at high relative humidity (RH) (Angellier-Coussy, Torres-Giner, Morel, Gontard, & Gastaldi, 2008). The mechanical properties of gluten-based films are strongly affected by pH and gluten concentration while water vapor permeability may be correlated with pH and ethanol level (Gontard, Duchez, Cuq, & Guilbert, 1994). Although the effects of plasticizers and hydrophobic constituents on properties of these

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films have not tested, the addition of these constituents is suggested to optimize the mechanical and moisture barrier properties. Fakhouri, Tanada-Palmu, and Grosso (2004), for example, studied composite films of wheat gluten and cellulose acetate phthalate and concluded that the composite films present more significant barrier properties to water than single films produced from the same materials. Curiously, in another study, it was showed that the addition of thyme oil as the active agent in gluten films by thermoplastic processing provide both antioxidant and antimicrobial properties. Although this constituent also leads to heterogeneous films with hydrophobic discontinuities that reduce the mechanical performance in terms of strength and modulus, at the same time, it enhances the flexibility of the film (Ansorena, Zubeldía, & Marcovich, 2016).

Gelatin is a product of collagen degradation, which involves the breakdown of the triple-helix structure into random coils (Kozlov & Burdygina, 1983) and its film-forming ability is well-established (Yakimets et al., 2007). Gelatin, which is able to form clear and strong films, is often used for microencapsulation and capsule coatings in both food and pharmaceutical industry (Lens et al., 2003). Bertan, Tanada-Palmu, Siani, and Grosso (2005) have studied the effect of fatty acids and 'Brazilian elemi' on composite films based on gelatin. Films containing fatty acids presented higher water vapor barrier properties than gelatin/triacetin films. The mechanical resistance was decreased by the addition of lipids while the opacity and soluble matter increased. Composite edible protein films combined with lipids can present more significant barrier properties than those films prepared only with proteins (Galus & Kadzińska, 2015, 2016; Otoni, Avena-Bustillos, Olsen, Bilbao-Sáinz, & McHugh, 2016).

Taking into account the advantages of the above materials, this study focused on the development of edible gluten-gelatin films containing plasticizers and fatty acids to improve the mechanical and water barrier properties. The fatty acids selected were caproic, caprylic, capric, lauric, myristic and palmitic in order to consider the hydrophobicity effect on film properties. Thermal and water barrier properties, solubility and opacity of the films were considered.

2. Materials and methods

2.1. Materials

Gelatin type A, wheat gluten, glycerol and sorbitol were obtained from Leiner Davis Gelatin, Rhodia, Synth and Getec, respectively (Brazilian suppliers). Caproic, caprylic, capric, lauric, myristic and palmitic fatty acids were obtained from Sigma-Aldrich (Brazil).

2.2. Film preparation and composition

Films were obtained by the casting solvent technique proposed by Gontard, Guilbert, and Cuq (1992) with minor modifications (citric acid was used to adjust the pH). Prior to the preparation of the protein blend that would form the films, gluten and gelatin solutions were prepared separately. After this step of solubilization of the protein and inclusion of the plasticizer, the solutions were mixed in the desired proportions so that the final formulations of each film may be obtained. GLY or SOR were used as plasticizer because they are edible, inexpensive and compatible with the film macromolecules (Vanin, Sobral, Menegalli, Carvalho, & Habitante, 2005).

The gluten solution contained 318.8 mL of distilled water, 431.2 mL of ethanol, 37.5 g or 75 g of gluten (5 or 10% solution of GLU, respectively → 5 or 10GLU10GEL) and 0.2 g plasticizer/g

gluten. The pH was adjusted to 3.0 by the addition of citric acid, and the mixture was heated at 70 °C under magnetic stirring. Subsequently, the gluten solution was centrifuged at 3500 rpm for 25 min. The liquid phase was separated from the solid (viscous) phase and stored for later use.

The gelatin solution was obtained in two steps. In the first step, 10 g of gelatin type A was hydrated in 100 mL of water. Afterward, this solution was heated at 70 °C for 10 min, and the plasticizer (sorbitol or glycerol) was included in a proportion of 0.05 g plasticizer/g gelatin.

To obtain the filmogenic solution, gluten and gelatin solutions were mixed on a heating plate at 60 °C under magnetic stirring at volume ratios of 1:1, 1:4 and 4:1 (gluten solution/gelatin solution). Films were obtained by pouring 20 mL of filmogenic solution into Petri dishes (11.8 cm diameter) and drying at 25 °C for 24 h. After drying, films were stored at 25 °C and 55± 2% relative humidity (RH) for at least 48 h prior to analysis. Films containing different plasticizer are summarized in Table 1. The composition of films containing fatty acids is 10GLU10GEL/SOR (1:1) with different FA amounts (5, 15, 25 and 50 g/100 g polymer).

Fatty acids were added to the solution under magnetic stirring (30 min, 200 rpm) at 60 °C. The mixture showed to be stable and no apparent phase separation in the films was observed. No exudation of fatty acids from the films was also observed during the drying process, probably due to an interaction of these constituents with the protein matrix, which involved hydrogen bonds and physical entanglement interactions.

2.3. Physicochemical characterization

2.3.1. Visual aspect and thickness

A visual trial was performed to select only homogeneous and flexible films (presenting a homogeneous color and no insoluble gelatin particles). Sample thickness was measured by using a micrometer (Model MDC-25 M, Mitutoyo, MFG, Japan). A minimum of 10 points was measured for each sample. After that, films were kept at 25 °C and 50% RH for 48 h before analysis.

2.3.2. Water and acid solubility

The water solubility assays were carried out according to (Gontard et al., 1994). The solubility was expressed as the percentage of dry matter of the film solubilized after a 24 h immersion in water. The solubility in acidic solution was determined in a chloride acid solution (1 mol/L) for 24 h. The measurements were performed in triplicate.

2.3.3. Film opacity

Film opacity was determined by using a HunterLab colorimeter (Colorquest II, Fairfax, USA). The calibration of the instrument was performed with black and white backgrounds. Opacity was determined according to the equation below (HUNTERLAB, 1997):

$$Op = \left(\frac{Op_N}{Op_B} \right) 100 \quad (1)$$

where: Op = film opacity (%), Op_N = film opacity on a black background, Op_B = film opacity on a white background.

2.3.4. Water vapor permeability (WVP)

The water vapor permeability (WVP) of films was measured according to a modified ASTM E96 standard method. Films were placed in aluminum cells containing calcium chloride (relative humidity close to 0.01% at 25 °C), and sealed with paraffin. Permeation cells were placed in desiccators maintained at 25 °C and 75% RH. The amount of water vapor migrating through the film

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