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Effect of airflow presence during the manufacturing of biodegradable films from polymers with different structural conformation

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ABSTRACT

The preparation method to obtain biodegradable films has an important effect on their performance. The drying of film-forming solutions is a very important step usually ignored during the manufacturing of biodegradable films. The objective of this study was to evaluate the effect of the drying process (natural convection or forced convection) on the functional properties of films from polymers with different structure. The film-forming solutions were dried using airflow (forced convection) and no air circulation (natural convection). Films from polyvinyl alcohol (PVOH, with linear structure) and polymers with different degree of branching (potato starch and high amylose corn starch) were evaluated. Films dried in forced convection showed important changes in the water adsorption, mechanical and thermal properties. Drying under forced convection could have promoted the formation of intermolecular bonds during the rearrangement of the polymer chains resulting in films with greater barrier to water vapor and better mechanical performance.

1. Introduction

Natural polymers have been widely used in food packaging due to its low toxicity, biocompatibility and biodegradability. Starch, the main polysaccharide used for storing energy in the vegetable kingdom, is abundant in nature and easily available as a commercial product. It is used widely in food packaging due to its low cost and good film-forming properties (Liu et al., 2017; Romero-Bastida, Bello-Pérez, Velazquez, & Alvarez-Ramirez, 2015). Starch is a mixture of amylose, a glucan predominantly linear and amylopectin, a high-molecular weight, highly branched glucan (Kramer, 2009). The amylose is more closely associated with the capability to form films and coatings, due to its predominantly linear nature. A film or coating can be made from any kind of starch containing amylose.

Starches with high content of amylose are obtained from genetically modified corn. However, the main disadvantage of these starches is a highly crystalline structure, requiring high temperatures to achieve a complete gelatinization (Menzel et al., 2015). Gelling the starch granules with high content of amylose until complete dispersion requires temperatures above 100 °C, which can only be achieved above the atmospheric pressure (Kramer, 2009). Starches with 50, 70 or even 90% amylose content have been used to obtain biodegradable films (Romero-Bastida et al., 2015; Wang, Wang, Yu, & Wang, 2014).

Polyvinyl alcohol (PVOH) is a synthetic polymer produced in high volume worldwide, because it has excellent physical properties, chemical resistance and complete biodegradability (Guirguis & Moselhey, 2012). PVOH has a linear structure; it is non-toxic and non-carcinogenic material with good compatibility with other materials, excellent film-forming capacity, high emulsifying capacity and excellent adhesive properties, which have resulted in extensive industrial uses. PVOH is considered as a synthetic truly biodegradable polymer approved by the FDA, so it can be used for food packaging applications (Buonocore et al., 2003; Curley, Hayes, Rowan, & Kennedy, 2014; Tang & Alavi, 2011). PVOH has been used to obtain coatings that act as moisture barrier in nutritional supplements and in foods that need to be protected from moisture adsorption (Ghaffari-Moghaddam & Eslahi, 2014). Due to the excellent mechanical properties, PVOH has been studied for biomedical applications (Alves et al., 2016; Carvalho et al., 2009; Curley et al., 2014; Zhang et al., 2015), as food packaging and carrier for antimicrobial agents (Buonocore et al., 2003; Han, Wang, Li, Lu, & Cui, 2014; Muppalla, Kanatt, Chawla, & Sharma, 2014). PVOH-based films have a crystalline structure reflected in a lower water adsorption

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capacity than other hydrophilic materials; possess high resistance to traction and flexibility, as well as excellent properties of barrier to oxygen and aromatic compounds (Mucha, Ludwiczak, & Kawinska, 2005; Tang & Alavi, 2011).

The process for manufacturing edible biodegradable films using the solution casting method plays a critical role in their final performance (Bourtoom & Chinnan, 2008; Romero-Bastida et al., 2015). Drying is a very important step as it has been reported that some properties of the films as the crystallinity, depend on the conditions of processing, source of the biopolymer, plasticizer type, order of addition of components, final moisture content and the drying conditions (air speed and temperature) (Aguirre-Loredo, Rodríguez-Hernández, Morales-Sánchez, Gómez-Aldapa, & Velazquez, 2016; Kramer, 2009; Ortiz, de Moraes, Vicente, Laurindo, & Mauri, 2017; Romero-Bastida et al., 2015).

The drying methods and processing conditions affect the properties and functionality of different edible films (Chiou et al., 2009; Denavi et al., 2009; Liu et al., 2016; Mayachiew, Devahastin, Mackey, & Niranjan, 2010; Oliveira de Moraes, Scheibe, Augusto, Carciofi, & Laurindo, 2015; Ortiz et al., 2017; Tapia-Blácido, Sobral, & Menegalli, 2013; Thakhiew, Devahastin, & Soponronnarit, 2010). In the drying process, time and temperature are the main parameters affecting the crystallinity and mechanical properties (Donhowe & Fennema, 1994). Fast drying of polymer solutions can result in fractures or ripples limiting the intermolecular associations within the structure of the material, since the mobility of polymeric chains is restricted after solvent evaporation. For example, microwave drying reduce the lumpiness and shine of edible films (Skurtys et al., 2010). Ortiz et al. (2017) found that soy protein films dried by IR radiation were thicker than those dried by conductive heat, due to a lower degree of compaction of the polymer matrix as a result of the cross-linking or molecular unfolding within the network of proteins in the film. The drying method and the temperature modified protein conformation and determined the capability of protein chains to interact with each other during film formation. The type and number of interactions involved in this process could determine the degree of crosslinking and they are also correlate with the final physicochemical, mechanical, and barrier properties of the films.

A sparsely reported parameter is the presence or absence of airflow during the drying process, and still less reported, is the speed of the airflow, which is a highly important parameter influencing the functional properties of packaging materials. The structural ordering could be promoted by the formation of intra and intermolecular links induced by the reorganization of the polymer chains (Angellier-Coussy, Gastaldi, Gontard, & Guillard, 2011). The internal structure can be assessed trough the crystallinity index as well as thermal and mechanical properties, since a reduction in crystallinity may be associated with a lower breaking resistance (less stress) and a more elastic behavior (Ziani, Oses, Coma, & Maté, 2008).

The objective of this study was to evaluate the effect of the presence of airflow during the drying process on the structural, mechanical performance and barrier properties of edible biodegradable films prepared from polymers of different structure: polyvinyl alcohol (PVOH), which has a linear structure, and starches with different ratio of linear (amylose) and branched (amylopectin) structures: potato starch with 30% amylose and high amylose corn starch (70% amylose).

2. Materials and methods

2.1. Materials

Polyvinyl alcohol (PVOH, Mw = 124000 - 186000 $g{\cdot}mol^{-1}$, 98–99% hydrolysis) was purchased from Aldrich Chemical Company, Inc. (Milwaukee, WI, USA). Native potato starch with 30% of amylose and 70% amylopectin, was obtained from KMC (Germany). Corn starch with a high content of amylose (70%) Hylon VII was obtained from Ingredion (Bridgewater, N.J., USA). Glycerol (purity > 90%), obtained from Meyer (Mexico), was used as plasticizing agent.

2.2. Preparation of film-forming solutions

Film-forming solutions of potato or corn starches, were prepared by dissolving 5 g of polymer in 100 mL of distilled water and added with 30% (w/w) of glycerol. The concentration of plasticizer was defined in a preliminary test, as the minimum amount necessary to obtain manageable and ductile films. Potato starch solution was gelatinized at 90 °C in a heating plate with constant agitation, while high amylose corn starch was gelatinized in autoclave at 121 °C for 15 min (Romero-Bastida et al., 2015). PVOH solution was prepared at 4% (w/v) and added with 25% (w/w) of glycerol, the mixture was hydrated at room temperature and subsequently the temperature was increased to 70 °C to achieve complete dissolution.

2.3. Drying conditions

Films were prepared using the casting method pouring 80 mL of polymeric solution in glass molds of 130 x 130 mm. All film-forming solutions were dried in an oven at 62.5 °C with natural or forced convection (Oven Series 9000, Thermolyne). The films dried under forced convection were dried during 4 h, the speed of the airflow was 1 m/s, measured with a digital anemometer (Peakmeter Instrument Co., model MS6252A, China) with a relative humidity of 54%. It took 6 h to dry the films under natural convection at the same temperature.

2.4. Films characterization

2.4.1. Thickness

Film thickness was measured just after unmolding, at room conditions (35–45 %RH and ~25 °C) using a digital micrometer (Mitutoyo, model C112EXB, USA, 0.001 mm precision). Measurements were performed in ten random points on the entire surface of the film, periphery and center. For subsequent calculations, the average value of the ten measurements in each film, was used.

2.4.2. Density

Density was calculated by dividing the dry weight of a film sample by the volume, which was calculated by multiplying the area of the sample by thickness. The average value and standard deviation of 7 samples were obtained for each polymer and for each drying process.

2.4.3. Solubility in water

Films were cut into 20 x 20 mm samples and dried at room temperature (~ 25 °C) for 7 days over silica gel, to remove as much water as possible, and weighed. The films were placed individually in tubes of 50 mL with plastic lid and added with 30 mL of distilled water. The tubes were closed and kept at room temperature for 3 h with intermittent agitation cycles. The content of the tube was filtered on filter paper and dried at 120 °C for 20 h to determine the dry matter. The total percentage of soluble material (%) was calculated using the Eq. (1) (Wang et al., 2017). Five replicates were performed for each sample.

$$Solubility\ (\%) = \left[\frac{(Dry\ \ initial\ \ weight - Dry\ \ matter\ \ weight)}{Dry\ \ initial\ \ weight}\right] \times 100 \quad (1)$$

2.4.4. Morphology of films

Scanning electron microscopy (SEM) was used to evaluate changes in the microscopic structure, the homogeneity of the film and the surface morphology of the materials as a function of the drying processes. Film samples were mounted on bronze stubs using a double-sided tape and coated with a layer of gold, allowing the visualization of the surface. All samples were examined using a voltage of 20 kV at 250X magnification.