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Research paper

Active and smart biodegradable packaging based on starch and natural extracts

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ABSTRACT

Active and smart biodegradable films from cassava starch and glycerol with 5 wt.% of different natural extracts such as green tea and basil were obtained by casting. Their functional capacity as antioxidants and their physicochemical properties achieved from the incorporation of these types of extracts were evaluated. The content of phenolic compounds in the extracts led to films with significant antioxidant activity, being greater in the case of the system containing green tea extract. Color changes in both materials after immersion in different media (acid and basic) due to the presence of chlorophyll and carotenoids in the extracts were observed, but the film with basil extract reacted most notably to the different pH. These films degraded in soil under two weeks and were thermal stable up to 240 °C. Finally, the incorporation of extracts of green tea and basil led to thermoplastic starch films with lower water vapor permeability retaining their flexibility.

1. Introduction

Contaminations associated with synthetic food packaging and current increasing concerns related to the negative environmental impact of plastic packaging materials derived from petroleum, have driven significant interest from both academia and industry in natural and biodegradable materials (Bonilla, Talón, Atarés, Vargas, & Chiralt, 2013; Chang-Bravo, López-Córdoba & Martino, 2014; Fama, Bittante, Sobral, Goyanes, & Gerschenson, 2010; Gonzalez Seligra, Medina Jaramillo, Famá, & Goyanes, 2016). Different authors studied the feasibility of using starch films as packaging of food products due to their easy manipulation and optimal properties for that application (Bonilla et al., 2013; Campos, Gerschenson, & Flores, 2011; Mali, Grossmann, García, Martino, & Zaritzky, 2004; Versino, Lopez, Garcia, & Zaritzky, 2016). However, the current demand of consumers for more durable food products makes necessary the development of eco-friendly materials that are also functional, such as packaging with active compounds that can improve the quality of the products they cover, further contributing to the nutritional value of food. In this sense, several researchers investigated the use of antioxidants (Cerruti et al., 2011; Moreno et al., 2015), or antimicrobial agents in polymer matrices (Pelissari, Grossmann, Yamashita, & Pineda, 2009), obtaining the so-called "functional packaging". Although today packaging are composed of multiple layers, the possibility of developing edible films with these capabilities as "primary packaging" (i.e. the layer in direct contact with food) is under investigation (Van Herpen, Immink, & Van den Puttelaar, 2016; Wu & Dunn, 1995).

On the other hand, there are also defined intelligent packaging; a packaging system that is capable of carrying out intelligent functions (such as detecting, sensing, recording, tracing, communicating, and applying scientific logic) to facilitate the decision to extend shelf life, enhance safety, improve quality, provide information and warn about possible problems (Kerry, O'Grady, & Hogan, 2006; Yam, Takhistov, & Miltz, 2005).

Changes in pH are one of the indicators of the state and the quality of a food product (Dainelli, Gontard, Spyropoulos, Zondervan-van den Beuken, & Tobback, 2008). In most cases, when deterioration occurs in a food product, a change in pH is observed. In this sense, pH evaluation is essential in a product before buying or consuming it (Bamore, Luthra, Mueller, Pressley, & Beckwith, 2003; Veiga-Santos, Ditchfield, & Tadini, 2011).

There are components such as chlorophyll and carotenoids, which possess the property of giving yellow-green pigmentation. These kinds of components can be found in green tea and basil, in particular in their extract form, and they undergo color changes when they are exposed to different pH conditions (Lee, 2012).

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Natural extracts are sources of antioxidants such as polyphenols and flavonoids, among others, whose activity is well known in pharmaceutical, cosmetic and food industries (Yilmaz & Toledo, 2006). Additionally, they could improve the plasticizing properties of biomaterials (Mathew, Brahmakumar, & Abraham, 2006; Medina Jaramillo, González Seligra, Goyanes, Bernal, & Famá, 2015).

The extract of green tea (*Camellia sinensis*), contains polyphenols that mainly include catechins, epicatechins, epigallocatechins, epicatechin gallate, epigallocatechin gallate, chlorogenic acid and gallic acid. The epigallocatechin gallate is the most abundant catechin that has major health benefits and prevents various diseases (Jiang, Engelhardt, Thräne, Maiwald, & Stark, 2015; Pasrija, Ezhilarasi, Indrani, & Anandharamakrishnan, 2015; Yang & Koo, 1997). "Perazzo et al. (2014) incorporated green tea extract into a starch matrix and reported significant improvements in the films mechanical properties, water vapor barrier properties and antioxidant activity. However, the ability of green tea extract as an active component in starch films to generate an intelligent coating has not been already investigated.

Basil (Ocimum basilicum L.) is an aromatic herb that has been traditionally used as a medicinal herb for the treatment of headaches, coughs, diarrhea, constipation, warts, worms and kidney malfunctions (Politeo, Jukic, & Milos, 2007; Simon, Morales, Phippen, Vieira, Hao, & Janick, 1999). It contains active compounds with antioxidant qualities such as linalool, chlorogenic acid and methyl chavicol, and it is stable at relatively high temperatures, being able to resist starch casting process to be used in packaging applications (Suppakul, Sonneveld, Bigger, & Miltz, 2008). These attractive characteristics of basil along with its ability to detect changes in the quality of a product and to impart pleasing sensorial characteristics to the consumer make this component very promising for using in the food packaging industry. However, to date, no studies have been found in the literature regarding the effect of basil on thermoplastic starch films, neither on its potential use as a component of active and intelligent packaging.

Migration evaluation of polymer based films is explained from the physical principle of diffusion and according to Article 2 of the European Economic Community (EEC) Directive 85/572/EEC, and it uses accelerated tests to estimate the real conditions avoiding the long analysis times (Baner, Bieber, Figge, Franz, & Piringer, 1992). According to the literature, water is a typical simulant of foods that have high water content and ethanol as a fatty food simulant, in migration tests (Busolo & Lagaron, 2015; Paseiro-Cerrato et al., 2017; Rodríguez-Martínez et al., 2016). In particular, Talón et al. (2017) developed starch-quitosan based films that presented water solubility of around 23% and investigated the kinetics of release of polyphenols from thyme extract in the films using different solvents as food simulants, including water (Talón, Trifkovic, Vargas, Chiralt & González-Martínez, 2017).

The aim of this work was to determine the ability of green tea and basil extracts to generate active and intelligent primary packaging from their antioxidant components and their pH-related color changes when incorporated into a biodegradable and edible starch based film.

2. Experimental

2.1. Materials

Cassava starch (18 wt.% amylose and 82 wt.% amylopectin) was provided by *CODIPSA, Paraguay*. Analytical grade glycerol (Aldrich) and commercial *yerba mate* (*Ilex paraguariensis*) (*Taragüi* liviana) from *Establecimiento Las Marías*, Corrientes, Argentina were used.

2.2. Preparation of green tea and basil extracts

Green tea and *basil* extracts were obtained using the infusion methodology previously described by Medina Jaramillo et al. (2015). Commercial green tea or basil leaves (~3 g) were immersed in 100 mL of distilled water and heated at 100 °C for 40 min. After that, the system were cooled, filtered through a mesh of approximately 20 μm, and stored in dark flasks until use.

2.3. Films formation

Three different thermoplastic starch films were prepared by casting according the methodology described by Medina Jaramillo et al. (2015). The thermoplastic starch (TPS) matrix consisted on cassava starch (5 wt.%), glycerol as plasticizer (1.5 wt.%) and distilled water (93.5 wt.%). The films with natural extracts (green tea, *TPS-T*, and basil, *TPS-B*) had the same concentration of starch and glycerol than the matrix but 5 wt.% of the distilled water was replaced by each extract. All components were mixed and homogenized for 45 min at ambient temperature (25 °C) with controlled stirring. Then, the mix was heated at 96 °C for 40 min to ensure starch gelatinization (Hernández, Emaldi, & Tovar, 2008; Medina Jaramillo et al., 2015). The gel was degassed by applying vacuum for 7 min and it was deposited on polypropylene plates, which were dried at 50 °C for 48 h at ambient relative humidity (RH). The thickness of the resultant films was ~0.25 mm.

2.4. Films characterization

All films were conditioned at RH of 56.7% and ambient temperature for 10 days before characterization.

2.4.1. Total polyphenols content (TPC)

Total polyphenols content (TPC) was determined by the Folin–Ciocalteu methodology (Singleton, Orthofer, & Lamuela-Raventós, 1999), as follow: 160 μL of Na_2CO_3 (7% w/v) (Anedra, Argentina) were mixed with 400 μL of the sample and 200 μL of Folin–Ciocalteau reagent (Anedra, Argentina, 1:10 diluted). After 30 min, sample absorbance was measured at 760 nm in a spectrophotometer (Shimadzu, UV-1800, Japan). Chlorogenic acid (Fluka, USA) was used as standard. Determination was performed in triplicate.

2.4.2. Migration tests of green tea and basil extract from active films to food simulants

Migration tests were performed following the methodology reported by Busolo and Lagaron (2015), Baner et al. (1992), and Article 2 of the EEC (European Economic Community Directive 85/572/EEC), which indicate that water can be used as aqueous simulant of foods. Samples (film pieces of ~ 200 mm^2) were deposited in 5 mL of water and placed in an orbital shaker at 25 °C at 100 rpm for 24 h. After that, migration of both green tea and basil polyphenols was evaluated by the Folin-ciocalteu method.

2.4.3. Scanning electron microscopy (SEM)

Cryo-fractured surfaces were examined using a scanning electron microscope with a Field Emission Gun (FEG) Zeiss DSM982 GEMINI, in order to investigate the morphology of the films.

Samples were cooled in liquid nitrogen, broken and coated with a thin sputtered gold layer before the analysis.

2.4.4. Water vapor permeability (WVP)

Water vapor permeability (WVP) tests were carried out following ASTM-E96-00, (1996) standard recommendations, and using the correction method described previously by (Gennadios, Weller, & Gooding, 1994). WVP ($g\,Pa^{-1}\,s^{-1}\,m^{-1}$) values were calculated as follows:

$$WVP = \left(\frac{G}{P \times RH \times A}\right) \times d \tag{1}$$

where G is the weight gain in time ($g\,s^{-1}$), A the cell area (m^2), P the saturation vapor pressure of water (Pa), RH the relative humidity, and d the film thickness (m).