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Biodegradability and plasticizing effect of *yerba mate* extract on cassava starch edible films

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A B S T R A C T

Biodegradable and edible cassava starch-glycerol based films with different concentrations of *yerba mate* extract (0, 5 and 20 wt.%) were prepared by casting. The plasticizing effect of *yerba mate* extract when it was incorporated into the matrix as an antioxidant was investigated. Thermal degradation and biodegradability of the obtained biofilms were also studied. Thermogravimetric analysis (TGA), differential scanning calorimetry (DSC), attenuated total reflectance Fourier transform infrared spectroscopy (ATR/FTIR), X-ray diffraction analysis (XRD), water absorbance, stability in different solutions and biodegradability studies were performed. The clear correlation among the results obtained from the different analysis confirmed the plasticizing effect of *yerba mate* extract on the starch-glycerol matrix. Also, the extract led to a decrease in the degradation time of the films in soil ensuring their complete biodegradability before two weeks and to films stability in acidic and alkaline media. The plasticizing effect of *yerba mate* extract makes it an attractive additive for starch films which will be used as packaging or coating; and its contribution to an earlier biodegradability will contribute to waste reduction.

1. Introduction

Packaging has a key role in containing and protecting food since it is highly manipulated by producers and consumers. However, packaging materials are one of the main solid wastes in major cities around the world (Elgaaïed-Gambier, 2014; Potter & Hotchkiss, 2012). A large portion of this waste is food containers. The reduction of the environmental impact by using biodegradable polymers in food industry is today an alternative for removing packaging from petroleum (Bonilla, Talón, Atarés, Vargas, & Chiralt, 2013; Chang-Bravo, López-Córdoba, & Martino, 2014; Seligra, Nuevo, Lamanna, & Famá, 2013). The term "biodegradable" materials is used to describe those materials which can be degraded by the enzymatic action of living organisms, such as bacteria, yeasts, fungi and the ultimate end-products of the degradation process, these being CO_2, H_2O, and biomass under aerobic conditions and hydrocarbons, methane and biomass under anaerobic conditions (Avella et al., 2005).

In this sense, starch edible films have been extensively studied in the last decades (Famá, Rojo, Bernal, & Goyanes, 2012; Gutiérrez, Morales, Pérez, Tapia, & Famá, 2015; Gutiérrez, Tapia, Pérez, & Famá, 2015b; López-de-Dicastillo, Gómez-Estaca, Catalá, Gavara, & Hernández-Muñoz, 2012; Machado, Nunes, Pereira, & Druzian, 2012; Souza et al., 2012).

Recently, it was demonstrated that starch-based products degradation time is shorter than other polymeric materials such as poly(lactic acid) (PLA) or poly(butylene adipate-*co*-terephthalate) (PBAT) (Weng et al., 2013). One of the advantages of a faster material degradation in landfill is that the volume of waste to be compacted would be also reduced. A decrease in the degradation time, even of a few days, represents a significant reduction of waste volume and hence, it leads to a very important benefit with regard to environmental care.

Several natural and synthetic additives are easily available for the development of starch based thermoplastics, such as process-

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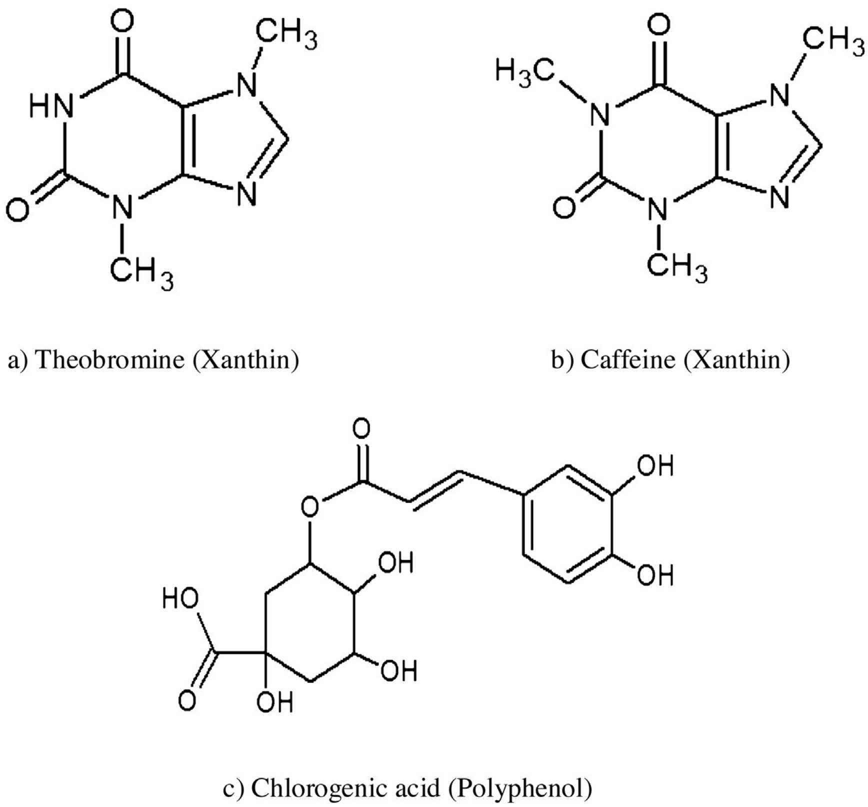

a) Theobromine (Xanthin) b) Caffeine (Xanthin)

c) Chlorogenic acid (Polyphenol)

Fig. 1. Chemical structure of chlorogenic acid (a), caffeine (b) and theobromin (c).

ing aids, plasticizers, stabilizers, antimicrobial and antioxidants (Calatayud et al., 2013; López-de-Dicastillo et al., 2012; Mathew and Abraham, 2008; Soroka, 2002).

The primary role of the plasticizers is to improve the flexibility and processability of polymers. It is known that plasticizers are low molecular weight materials. This means they can occupy intermolecular spaces between the polymer chains, reducing secondary forces among them. These molecules change the three-dimensional molecular organization of polymers, reducing the energy required for molecular motion and the formation of hydrogen bonding between the chains (Vieira, da Silva, dos Santos, & Beppu, 2011). Thus, the degree of plasticity of polymers is largely dependent on the chemical structure of the plasticizer, including chemical composition, molecular weight and functional groups (Moreno, 1992).

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 and Toledo, 2006). They also are chemical analogues of synthetic stabilizers widely used in plastic industries (Cerruti, Malinconico, Rychly, Matisova-Rychla, & Carfagna, 2009). Additionally, the incorporation of natural extracts could improve the plasticizing properties of biomaterials (Mathew and Dufresne, 2002; Mathew and Abraham, 2008; Medina Jaramillo, González Seligra, Goyanes, Bernal, & Famá, 2015).

The composition of *yerba mate* has been partially characterized in the literature. It includes a variety of active phytochemicals, among them, the highest compounds are the polyphenols (chlorogenic acid) and xanthines (caffeine and theobromine) (Fig. 1), followed by purine alkaloids (caffeic acid, 3,4-dicaffeoylquinic acid, 3,5-dicaffeoylquinic acid), flavonoids (quercetin, kaempferol, and rutin), amino acids, minerals (P, Fe, and Ca) and vitamins (C, B1, and B2). These are the main constituents that are relevant for the antioxidant capacity of *yerba mate* (Bastos et al., 2007; Bravo, Goya, & Lecumberri, 2007; Filip, López, Giberti, Coussio, & Ferraro, 2001; Gosmann, Schenkel, & Selignmann, 1989; Pomilio, Trajtemberg, & Vitale, 2002).

The addition of *yerba mate* extract in biodegradable biopolymers could allow obtaining active films with antioxidant effects thus, having important benefits on the health of consumers (Bastos et al., 2007; Bravo et al., 2007; Filip et al., 2001; Gosmann et al., 1989; Pomilio et al., 2002). The effectiveness of yerba mate as an antioxidant when it was incorporated in a thermoplastic starch film (TPS) has been previously reported (Medina Jaramillo et al., 2015). Through DPPH• tests, the authors observed that the antioxidant activity of TPS containing 5wt.% and 20wt.% of extract of yerba mate was 3.0 ± 0.1 and 10.4 ± 0.1 mg/g of film, respectively.

Beyond that packaging can provide extra benefits to the food it must fulfill certain conditions to avoid adversely effects on the product or the environment. For this, some studies of thermal degradation reported that the temperature that can support a material, as well as its biodegradability in soil, should be fundamental in evaluating its ability to enter as packaging in market food industries. Biodegradation tests under composting conditions are important because they simulate the biodegradability under real life conditions and they are also treatment methods for municipal solid waste (Briassoulis, Dejean, & Picuno, 2010; Maiti, Ray, & Mitra, 2012).

In this context, some researchers studied the thermal degradation properties of starch-based films containing antioxidants (Moreno, Atarés, & Chiralt, 2015; Perazzo et al., 2014). In most investigations it has been shown that the addition of antioxidants leads to slight changes in thermal degradation parameters. Mainly, a beginning of material degradation at lower temperatures was reported. However, there are not researches that have investigated the effect of using an antioxidant in the degradation time in soil of functional packaging films with food oxidation inhibitors characteristics.