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Development of biocomposite films incorporated with different amounts of shellac, emulsifier, and surfactant

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ABSTRACT

This study examined the effects of different ratios of shellac (20–60%), stearic acid (SA) (0–2%), and Tween-20 (0.1–0.5 ml) on the water vapor permeability (WVP) and mechanical properties of the pea starch-guar gum (PSGG) films which were evaluated by using response surface methodology (RSM). The incorporation of shellac into the PSGG film structure led to a slightly increased of film thickness. However the addition of higher concentrations of shellac did not improve the moisture barrier of PSGG film owing to the poor distribution of shellac in the film structure. Film formulated with 40% shellac, 1% SA, and 0.3% Tween-20 exhibited optimal functional properties. Moreover, the influence of the incorporation of different emulsifiers into the optimized film matrix was investigated by studying the physical, mechanical, and optical properties of the films. Films containing oleic acid (OA) showed not only lower thickness, WVP, moisture content, and water solubility, but also higher percentage of elongation (E), tensile strength (TS), and transparency compared with other fatty acids tested. Biocomposite pea starch-guar gum-shellac (PSGG-Sh) films containing OA can be considered to be sufficient for most of food packaging applications.

1. Introduction

Safety and quality properties of food products which incorporate chemical changes (enzymatic browning and oxidation), microbial stability, sensorial (texture) and physical characteristics, determine the product quality and shelf life (Phan The, Debeaufort, Luu, & Voilley, 2008). The application of edible films and coatings to reduce deteriorative processes, as well as increasing shelf-life and appearance of food, has attracted significant research and industry interest (Bosquez-Molina, Guerrero-Legarreta, & Vernon-Carter, 2003). Edible films made from starch are suitable for food preservation because their resultant films are odorless, tasteless, and transparent with good oxygen barrier properties. Although starch based films have some limitations due to their affinity to water adsorption and retrogradation phenomena which affect their mechanical and barrier properties of films (Cano, Jimenez, Chafer, Gonzalez, & Chiralt, 2014; Jiménez, Fabra, Talens, & Chiralt, 2013). One approach to overcome these shortcomings is development of composites with other polymers or reinforcement substances (Ortega-Toro, Jiménez, Talens, & Chiralt, 2014).

Guar gum (GG) derived from a legume plant *Cyamopsis tetragolonoba* is an appropriate biopolymer for formation of biodegradable films owing to its high molecular weight and wide availability (Saurabh, Gupta, Variyar, & Sharma, 2016). GG is a galactomannan with a backbone of 1, 4-linked β-D-mannose residues and galactose as a side group linked by (1–6) α-d-galactopyranose at every second mannose which establishes short side-branches (Fernandes, Gonçalves, & Doublier, 1993).

Our previous studies demonstrated that GG in combination with pea starch (PS) improved physical, barrier and mechanical properties of films (Saberi et al., 2016a, 2017). The water vapor permeability (WVP) value of pea starch-guar gum (PSGG) biocomposite films exhibited better moisture barrier characteristics than pure PS films (Saberi et al., 2016b), but these were higher than those of low density polyethylene (LDPE) (Phan The et al., 2008). GG improved

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the functional characteristics of PS edible film as both biopolymers are hydrophilic; however, the incorporation of hydrophobic substances for decreasing the water sensitivity of biocomposite films is required.

Lipid components including natural waxes, fatty acids, essential oils, surfactants and resins are commonly applied to reduce water vapor transmission rate in the hydrocolloid matrix (Villalobos, Hernández-Muñoz, & Chiralt, 2006). Shellac is currently used as a moisture barrier in the food industry to extend the shelf-life of products (Phan The et al., 2008), and in the pharmaceutical industry for the moisture protection of drugs, controlled drug delivery system and as an enteric coating for drugs and probiotics (Pearnchob, Dashevsky, & Bodmeier, 2004; Soradech, Limatvapirat, & Luangtana-anan, 2013; Stummer et al., 2010). Shellac is a purified resinous secretion of lac insects, *Kerria lacca*, a parasitic insect found on trees in Southeast Asia (Phan The et al., 2008). However issues related to the application of shellac include its insolubility in an aqueous system, lack of mechanical strength and lower stability, which lead to the reduction in its use (Limmatvapirat, Limmatvapirat, Puttipipatkhachorn, Nuntanid, & Luangtana-Anan, 2007; Luangtana-anan et al., 2007). The production of composite films by combination of two natural polymers and the incorporation of some plasticizers, is a novel approach to counter these issues (Soradech et al., 2013).

In addition, the incorporation of fatty acids has been used to decrease water transmission through edible films. Fatty acids are polar lipids and their chain length and unsaturation degree has a significant effect on film properties (Fernández, de Apodaca, Cebrián, Villarán, & Maté, 2007). Other hydrophobic compounds with the potential to improve film characteristics are surfactants. These compounds, such as Tween-20, are amphiphilic substances, which are necessary for preparation emulsion films with suitable properties (Tongnuanchan, Benjakul, & Prodpran, 2014).

The widespread availability and low cost of PS and GG make the use of these compounds ideal for film formation and in combination with shellac as a hydrophobic substance make the combination of these compounds ideal in the development of packaging films. However it is critical to understand the interaction and relationships between these compounds to optimize film properties. In this study, the influence of different amounts of shellac, surfactant and emulsifier based on dry film matter on mechanical (tensile strength and percent of elongation at break) and barrier properties (WVP) of PSGG based films was investigated using response surface methodology (RSM). In addition, different emulsifiers were added to PSGG-Sh composite film based on optimized amount to determine the appropriate emulsifier to make films with improved mechanical, water vapor barrier and optical properties.

2. Materials and methods

2.1. Materials

Canadian non-GMO yellow pea starch with 13.2% moisture, 0.2% protein, 0.5% fat, 0.3% ash, and 36.25 ± 0.32% amylose was used in all experiments (supplied by Yantai Shuangta Food Co., Jinling Town, China). Guar gum (E-412) was purchased from The Melbourne Food Ingredient Depot, Brunswick East, Melbourne, Australia. Food grade shellac was purchased from Castle Chemicals (castlechem.com.au), NSW, Australia. Stearic acid (SA), lauric acid (LA), oleic acid (OA), butyric acid (BA), palmitic acid (PA) and Tween-20 were obtained from Sigma Aldrich, Australia. Glycerol was from Ajax Finechem Pty. Ltd, Australia and used as a plasticizer. All other chemicals were purchased from Merck Millipore, Pty., VIC, Melbourne, Australia.

2.2. Emulsion preparation

Optimized amounts of pea starch (2.5 g), guar gum (0.3 g) and 25% w/w glycerol based on the dry film matter were dissolved in 100 ml degassed deionized water with gentle heating (about 40 °C) and magnetic stirring. In another study, we determined the optimized amount of film components by using Box–Behnken response surface design (BBD) (Saberi et al., 2016a). The PSGG-Sh composite mixtures without emulsifier did not form a film, therefore, SA, was used to stabilize the PSGG-Sh composite films. Melted SA (0–2% db) and Tween-20 (0.1–0.5% wb) were added to the PSGG solution and the aqueous suspension was gelatinized at 90 °C for 20 min on a hot plate with continuous stirring. According to preliminary moisture barrier and mechanical tests (data are not shown), shellac was added the PSGG-SA-Tween 20-glycerol mixture at three different levels (20%, 40% and 60% db). Once the lipids were melted, samples were homogenized for 4 min at 22000 rpm using a T25 Ultra-Turrax (Ika, Staufen, Germany). After homogenization, the film solution was cooled to room temperature with mild magnetic stirring for 1 h to decrease air bubbles. Filmogenic suspensions (20 g) were cast onto Petri dishes (10 cm in diameter) and dried at 40 °C in an oven until reaching constant weight (about 24 h). Films were carefully peeled-off from Petri dishes and conditioned at 25 °C, 65% relative humidity (RH) for 72 h prior to further testing (Saberi et al., 2017).

In a second experiment, six film formulations were prepared with and without incorporation of emulsifiers (LA, OA, BA, and PA) with the same amount of the optimized level of SA. All the films were prepared with the same procedure described before. Concentration of shellac and Tween-20 were kept constant throughout the study.

2.3. Experimental design

The effect of process parameters (shellac (X1): 20–60%, SA (X2): 0–2%, and Tween-20 (X3): 0.1–0.5 ml)) on film mechanical and barrier properties was studied by applying a three-level-three-factor, Box–Behnken response surface design (BBD) with three central point replicates. All experimental runs are listed in Table 1. A second-order polynomial model was used to fit the experimental data obtained from the seventeen experimental runs:

$$Y = \beta_0 + \sum_{i=1}^{k} \beta_i X_i + \sum_{i=1}^{k-1} \sum_{j=2}^{k} \beta_{ij} X_i X_j + \sum_{i=1}^{k} \beta_{ii} X_i^2 + e_i \quad (1)$$

where various X_i values are independent variables affecting the responses Y; β_0, β_i, β_{ii}, and β_{ij} are the regression coefficients for intercept, interaction coefficients of linear, quadratic and the second-order terms, respectively and k is the number of variables (Saberi et al., 2017).

2.4. Film characterization

2.4.1. Thickness

A digital micro-meter (Mitutoyo, Co., Code No. 543-551-1, Model ID-F125, 139 Japan; sensitivity = 0.001 mm) was used to measure film thickness. The mean value from 10 different points for each film samples was measured (Fakhouri, Fontes, Innocentini-Mei, & Collares-Queiroz, 2009).

2.4.2. Water vapor permeability

Water vapor permeability (WVP) of films was examined gravimetrically using the method explained by Sun, Wang, Kadouh, and Zhou (2014) with some modifications. The films were sealed onto