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Application of biocomposite edible coatings based on pea starch and guar gum on quality, storability and shelf life of 'Valencia' oranges



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ABSTRACT

Novel edible composite coatings based on pea starch and guar gum (PSGG), PSGG blended with lipid mixture containing the hydrophobic compounds shellac and oleic acid (PSGG-Sh), and a layer-by-layer (LBL) approach (PSGG as an internal layer and shellac as an external layer), were investigated and compared with a commercial wax (CW) and uncoated fruit on postharvest quality of 'Valencia' oranges held for up to four weeks at 20 °C and 5 °C with an additional storage for 7 d at 20 °C. The incorporation of lipid compounds into the PSGG coatings (PSGG-Sh) generally resulted in the best performance in reducing fruit respiration rate, ethylene production, weight and firmness loss, peel pitting, and fruit decay rate of the coated oranges. Fruit coated with PSGG-Sh and a single layer PSGG coatings generally resulted in higher scores for overall flavor and freshness after four weeks at 5 °C followed by one week at 20 °C than uncoated fruit, as assessed by a sensory panel. Although the LBL coating reduced weight loss and respiration rate with improved firmness retention to a greater extent than the single layer PSGG coating, the bilayer coating also resulted in higher levels of ethanol causing increased perception of off-flavors. Overall results suggested that PSGG-based edible coatings could be a beneficial substitute to common commercial waxes for maintaining quality and storability, as well as extending shelf life of citrus fruit and potentially other fresh horticultural produce.

1. Introduction

Edible films and coatings are widely used to maintain the quality and shelf life of many horticultural products, including citrus (Baldwin et al., 2011). Edible films and coatings act as semi-permeable membranes which restrict the movement of gases and water vapor to reduce the rate of respiration and water loss from the fruit. Many films/coatings due to their barrier and mechanical properties can reduce the rate of physiological postharvest degradation (Baldwin, 1994; Baldwin et al., 1995; Park, 1999).

In many countries, harvested citrus fruit are commonly waxed during their processing and packing. This is to replace the natural wax which is damaged/removed with commercial harvesting, handling, processing and packing (Valencia-Chamorro et al., 2010). The commercial application of waxes not only reduces weight loss and shrinkage, but also enhances shine by increasing gloss (Rojas-Argudo et al., 2009). However, some waxes have been shown to negatively alter the internal atmosphere of the fruit by inducing anaerobic off-flavor development with the restriction of respiratory gas exchange (Martínez-Jávega et al., 1989).

Many modern citrus waxes are made of shellac (derived from the lac bug, *Kerria lacca*) or carnauba (derived from the leaves of the carnauba palm, *Copernicia prunifera*). However there is a need to improve the efficiency and sustainability of waxes applied to citrus.

Readily sourced and inexpensive coating materials which are effective at maintaining fruit quality during storage and shelf life are required. Pea (*Pisum sativum*) is widely grown around the world and contains 22–45% starch as the most plentiful carbohydrate in the seed (Hoover and Sosulski, 1991). Pea starch (PS) is comprised of a mixture of two homopolymers; a linear fraction, amylose, and a highly branched fraction, amylopectin. They are made of units of D-glucose with only two types of chain linkages, an α -(1 \rightarrow 4) of the main chain and an α -(1 \rightarrow 6) of the branch chains (Liu, 2005). Pea starch has high content of amylose; therefore it is a potential option for the production of starch-based edible films (Van Soest et al., 2002). Guar gum (GG), which is derived from the endosperm of the guar bean (*Cyamopsis tetragonoloba*),

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is a type of linear galactomannan with ratio of mannose to galactose units of 2:1 (Prajapat and Gogate, 2015). The molecular structure of guar gum is composed of $\beta(1 \rightarrow 4)$ -linked mannopyranose backbone, with several branch points from the C-6 position of mannopyranose, linked by $\alpha(1 \rightarrow 6)$ bond to a single D-galactopyranose sugar (Whistler and BeMiller, 1993). Owing to the long polymeric chain, high molecular weight and wide availability of pea starch and guar gum, they can be potential alternatives for production of renewable source based biodegradable edible coatings or packaging materials. In our previous studies, it has been shown that pea starch in combination with guar gum can form biocomposite edible films with preferable physical, optical and mechanical properties (Saberi et al., 2016a, 2016b, 2016c). However, edible coatings based on pea starch and guar gum have not been comprehensively explored as fruit coatings.

Due to the hydrophilic nature of pea starch-guar gum (PSGG) film, it is necessary to add a hydrophobic substance for decreasing the water sensitivity of the film. In this experiment, shellac (Sh) was added as a resin-based hydrophobic substance to increase its capability in increasing gloss and decrease water loss (Arnon et al., 2015). However, an issue with shellac films is their lack of permeability to gases, which results in the accumulation of ethanol and carbon dioxide (CO_2) and the development of off-flavors during storage (Baldwin et al., 1995; Dhall, 2013; Porat et al., 2005).

In this study, we investigated the influence of pea starch-guar gum (PSGG), pea starch-guar gum-shellac (PSGG-Sh), and PSGG/Sh bilayer composite coating, formed by first applying PSGG and then shellac (Sh) compared with fruit coated with commercial wax and uncoated fruit (control) on maintaining the quality of fresh 'Valencia' oranges during four weeks at 20 °C and four weeks of storage at 5 °C followed by one week at 20 °C, simulating marketing shelf life.

2. Materials and methods

2.1. Materials

Canadian non-GMO (non-genetically modified organism) yellow pea starch with 13.2% moisture, 0.2% protein, 0.5% fat, 0.3% ash, and 36.25 \pm 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 alcohol-based solution of shellac and Citrus Gleam (a shellac-based commercial wax) were purchased from Castle Chemicals (castlechem.com.au), NSW, Australia. Oleic acid (OA) 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. Sample preparation

'Valencia' oranges (*Citrus sinensis* L. Osbeck) were obtained from a local commercial citrus grower (Griffith, NSW, Australia) at commercial maturity and transported to the NSW Department of Primary Industries (Ourimbah, NSW, Australia). Oranges were selected based on homogeneity in shape, color, size, firmness and free of mechanical wounds or fungal decay. Selected oranges were dipped in a solution of 1150 μ L L⁻¹ fludioxonil (Scholar^{*}, Syngenta Australia) for one min, then drained and air-dried at 20 °C before coating application.

2.3. Coating formulations

2.3.1. PSGG coatings

Pea starch (2.5 g), guar gum (0.3 g) and 25% w/w glycerol as plasticizer based on the dry film matter were dissolved in 100 mL degassed deionized water. The solution was heated at 90 $^{\circ}$ C for 20 min upon constant stirring. The suspension was then cooled until room

temperature with mild magnetic stirring (Saberi et al., 2016b). The film solution was prepared one day before use.

2.3.2. PSGG-Sh coatings

The PSGG-Sh composite mixture was prepared by adding oleic acid (1% of dry weight of pea starch and guar gum) as emulsifier and Tween-20 (0.3 mL) as surfactant to the PSGG solution made as described above. Food grade alcohol-based solution of shellac at 40% (dry weight of pea starch and guar gum) was added to the PSGG-OA-Tween 20-glycerol mixture. These levels of film ingredients were optimized using Box–Behnken response surface design (Saberi et al., 2017). The emulsion was gelatinized at 90 °C for 20 min on a hot plate with continuous stirring. Once the lipids had 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 slow magnetic stirring. The emulsion was prepared one day before use and was shown to be stable with no phase separation.

2.4. Experimental design

Five series of treatments were applied on oranges: (i) PSGG; (ii) PSGG-Sh; (iii) bilayer formulation of PSGG as an inner layer with Sh solution as an external layer (PSGG/Sh); (iv) CW (commercial wax, shellac based 'Citrus Gleam') and (v) distilled water acting as a control. Each treatment for each storage condition included 128 oranges with 8 oranges per plastic netted bag. There were four replicates per treatment with each bag considered a single replicate. Data were recorded before treatment (day 0) and at 7 d intervals (four removals) for up to four weeks storage at 20 °C and relative humidity (RH) of 90–95%, and logging the temperature and RH with calibrated TinyTag View 2 loggers. Another set of treated oranges was also stored for 1, 2, 3, and 4 weeks at 5 °C and 90–95% RH, followed by one additional week at 20 °C to simulate retail handling and marketing conditions.

2.5. Fruit coating

Each coating solution was sprayed uniformly on the whole fruit surface by using a paint sprayer (High Volume Low Pressure system, 500W Paint Sprayer, 909, Mooroolbark, Vic, Australia). The bilayer coatings were applied as follows: first the PSGG coating was applied and fruit were fan dried at room temperature for 2–3 min and then the Sh coating was applied. Then, all coated oranges were air-dried for 1 h at 20 °C, labelled, weighed, and then randomly packed into experimental units. Fruits were destructively measured each week for up to four weeks at either 20 °C or 5 °C. Four oranges from each replicate were assessed upon removal (when the fruit had reached room temperature) and the remaining four fruit were stored for the additional week at 20 °C.

2.6. Fruit quality parameters

2.6.1. Weight loss

Fruit weight loss was measured by weighing the same marked fruit, at the beginning of the experiment and at the end of each storage period. The results were presented as the percentage loss of initial weight (Rojas-Argudo et al., 2009).

2.6.2. Fruit firmness

A texture analyzer (Lloyd Instrument LTD, Fareham, UK) was used to determine firmness of fruit upon each removal. The maximum force (N) was measured by compressing the fruit in the equatorial zone between two flat surfaces closing together at the rate of 1 mm min⁻¹ to a depth of 2 mm. The average of two reading points from each side of the fruit were recorded (Cháfer et al., 2012).

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