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Development of active and intelligent packaging by incorporating betalains from red pitaya (*Hylocereus polyrhizus*) peel into starch/polyvinyl alcohol films

Yan Qin, Yunpeng Liu, Xin Zhang, Jun Liu *

College of Food Science and Engineering, Yangzhou University, Yangzhou, 225127, PR China

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ABSTRACT

This study aims to develop active and intelligent packaging films based on starch/polyvinyl alcohol incorporated with betalains-rich red pitaya (*Hylocereus polyrhizus*) peel extract. The microstructure, physical and functional properties of the films with different contents of the extract (0.25, 0.50 and 1.00 wt% on starch basis) were measured. Results showed betacyanins were the main components in the extract, which presented significant color changes under alkaline conditions. Scanning electron microscopy observation revealed the compactness of the films was improved by the extract addition. Fourier transform infrared spectroscopy revealed that betacyanins in the extract could interact with starch, polyvinyl alcohol and glycerol through hydrogen bonds. The incorporation of the extract enhanced the water vapor barrier and ultraviolet–visible light barrier ability, and mechanical, antioxidant and antimicrobial potential of the films. The film containing 1.00 wt% of the extract was more sensitive to ammonia than other films. When applied to monitor the freshness of shrimp, the film containing 1.00 wt% of the extract presented visible color changes due to the accumulated volatile nitrogen compounds during the spoiling process of shrimp. Our results suggest the film containing 1.00 wt% of the extract can be used as active and intelligent packaging in food industry.

1. Introduction

The non-biodegradable synthetic polymer-based packaging has caused serious environmental problems (Otoni et al., 2017). So the development of natural and biopolymer-based packaging has become a global hotspot in recent years (Garavand, Rouhi, Razavi, Cacciotti, & Mohammadi, 2017). Starch, a natural and renewable biopolymer derived from numerous plant resources, has received increasing attention because it is abundant, cheap, biodegradable and edible (Cazón, Velazquez, Ramírez, & Vázquez, 2017). Starch also possesses good film-forming property and starch films are generally odorless, tasteless, colorless, non-toxic and biodegradable (Khan, Niazi, Samin, & Jahan, 2017). Nevertheless, pure starch films are highly hydrophilic and possess poor mechanical properties, thereby limiting their applications in food packaging (Cazón et al., 2017).

A promising approach to improve the physical properties of starch films is to blend starch with other polymers (Molavi, Behfar, Shariati, Kaviani, & Atarod, 2015). Polyvinyl alcohol (PVA), one of the water-soluble polymers, has been widely used in packaging materials due to its biodegradable, chemical resistant, film-forming, oxygen barrier and mechanical properties (Halima, 2016). PVA can be blended with starch to develop films with good mechanical and water resistant properties (Khan et al., 2017). However, starch/PVA films do not possess enough functional properties (e.g. antioxidant and antimicrobial activities) that are required for active packaging. In recent years, natural pigments including anthocyanins, betalains and curcumin have been added into starch-based films to improve the antioxidant and antimicrobial properties of the films (Liu et al., 2018, 2017a; Tran, Athanassiou, Basit, & Bayer, 2017). The developed active packaging films can be used to extend the shelf life of food products (Domínguez et al., 2018). Moreover, the films containing natural pigments are pH-sensitive and can be used as intelligent packaging to monitor the freshness of meat, milk and seafood (Liu et al., 2017a; Zhai et al., 2017; Zhang et al., 2019a). However, most existing studies have focused on the development of active and intelligent packaging by using anthocyanins from different sources.

Betalains are a family of water-soluble pigments produced by plants, including red beetroot, amaranth, prickly pear and red pitaya

* Corresponding author.
E-mail address: junliu@yzu.edu.cn (J. Liu).

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(Esatbeyoglu, Wagner, Schini-Kerth, & Rimbach, 2015; Polturak & Aharoni, 2019). Nowadays, betalains have been widely used as colorants in food industry. Structurally speaking, betalains can be sub-divided into red/violet betacyanins and yellow/orange betaxanthins (Gengatharan, Dykes, & Choo, 2015). As shown in Fig. 1, betacyanins are composed of betalamic acid (the chromophore) and cyclo-3,4-dihydroxyphenylalanine (cyclo-Dopa), whereas betaxanthins are the condensation of betalamic acid and amino acids or amines (Khan, 2016). Notably, betalains are structurally stable in the pH range of 3–7, but can undergo structural and color variations (from red to yellow) at alkaline conditions (Miguel, 2018). In recent years, several *in vitro* and *in vivo* experiments have demonstrated betalains possess antioxidant, antimicrobial, anti-cancer, anti-lipidemic and antidiabetic activities (Gandía-Herrero, Escribano, & García-Carmona, 2016; Gengatharan et al., 2015). Considering the superior antioxidant, antimicrobial and pH-sensitive properties, it is supposed that betalains can be used to fabricate active and intelligent packaging. Till now, studies about the development of active and intelligent packaging based on betalains are very limited (Ardiyansyah et al., 2018; Gutiérrez, Guzmán, Jaramillo, & Famá, 2016a; Jamróz, Kulawik, Guzik, & Duda, 2019).

Red pitaya (*Hylocereus polyrhizus*) is a vine fruit that belongs to the Cactaceae family (Tenore, Novellino, & Basile, 2012). The peel of red pitaya, representing approximately 18–24 wt% of the whole fruit, is usually discarded in practice. However, the peel of red pitaya contains abundant betalains (Wybraniec et al., 2001). Thus, recycling of betalains from the peel of red pitaya can create a higher economic value (Chew, Hung, & King, 2019). In this study, betalains were extracted from the peel of red pitaya and then incorporated into starch/PVA films to develop active and intelligent packaging. The effect of betalains content on the physical and functional properties of the films was evaluated. Meanwhile, the developed films were also applied to monitor the freshness of shrimp.

2. Materials and methods

2.1. Materials and reagents

Fresh red pitaya (*H. polyrhizus*) and shrimp was purchased from local market (Yangzhou, China). D101 macroporous resin was provided by Bohong Resin Technology Co., Ltd. (Tianjin, China). Cassava starch was obtained from Ganzhiyuan Sugar Industry Co., Ltd. (Nanjing, China). 2,2-Diphenyl-1-picrylhydrazyl (DPPH) and polyvinyl alcohol 1799 (degree of polymerization: 1750 ± 50, degree of alcoholysis: 98–100%) were purchased from Macklin Biochemical Co., Ltd. (Shanghai, China). Food-borne pathogens (*Escherichia coli*, *Staphylococcus aureus*, *Salmonella* and *Listeria monocytogenes*) were supplied by the Food Microbial Laboratory of Yangzhou University (Yangzhou, China).

2.2. Extraction of betalains

The peel of fresh red pitaya (300 g) was minced and extracted twice with 800 mL of 30% ethanol solution at 4 °C overnight (Azeredo, 2009). The extraction solution was filtered through a 200-mesh sieve and then centrifuged at 8000 *g* and 4 °C for 15 min to remove insoluble substances. The obtained supernatant was further purified on the column of D101 macroporous resin, which was first eluted with distilled water to remove water-soluble impurities (e.g. pectin and sugar) and then eluted with 30% ethanol solution to collect betalain-rich fraction. The eluate was concentrated and vacuum dried to afford red pitaya peel extract (RPPE). The extraction yield of RPPE was 0.32 g/100 g fresh peel. The prepared RPPE sample was stored at −20 °C and in darkness.

2.3. Characterization of betalains

2.3.1. Total betacyanins content

The total betacyanins content in RPPE was determined by the method of Mahayothee, Komonsing, Khuwijitjaru, Nagle, and Müller (2019). Briefly, RPPE was dissolved in McIlvaine buffer (pH 6.5) and the concentration of RPPE solution was 0.05 g/mL. After the solution was placed in the dark for 30 min, the absorbance of the solution was determined at 538 nm. The total betacyanins content was calculated by the following equation and expressed as mg betanin equivalents per g of dried extract.

$$\text{Betacyanin content (mg/g)} = \frac{\text{DF} \times A \times V \times \text{MW} \times 1000}{\varepsilon \times L \times M} \tag{1}$$

where DF is dilution factor, A is the absorbance at 538 nm, V is the volume of the solution (mL), L is path length of cuvette (1 cm), M is the weight of the extract (g). MW (550) and ε (65,000) represent the molecular weight and molar extinction coefficient of betanin, respectively.

2.3.2. pH-sensitive property

The pH-sensitive property of RPPE was determined by dissolving 2 mg of dried extract in 20 mL of buffer solutions (pH 3–12) for 30 min. The color of the solutions was captured by a digital camera and the visible absorption spectrum of the solutions was recorded on an ultraviolet–visible (UV–vis) spectrophotometer (Lambda 35, PerkinElmer Inc., MA, USA) from 400 to 700 nm (Jamróz et al., 2019).

2.4. Development of the films

Starch/PVA films with and without RPPE were prepared according to the method of Zhai et al. (2017) with some modifications. Briefly, starch (6.8 g) and PVA (3.4 g) were completely dissolved in 200 mL of distilled water at 100 °C with constant stirring for 90 min. Then, different contents of RPPE (0, 0.25, 0.50 and 1.00 wt%) on starch basis were added into the cooled starch/PVA solutions and stirred at 30 °C for 90 min. Afterwards, glycerol (25 wt% on starch basis) was added to obtain film-forming solutions. The solutions were spread on Plexiglas plates (24 cm × 24 cm) and dried at 30 °C for 48 h to afford the films. Starch/PVA films containing 0, 0.25, 0.50 and 1.00 wt% of RPPE were expressed as SP, SPR-0.25, SPR-0.5 and SPR-1.0 films, respectively. All the films were stored at 25 °C and 50% relative humidity (RH) for 48 h before test.

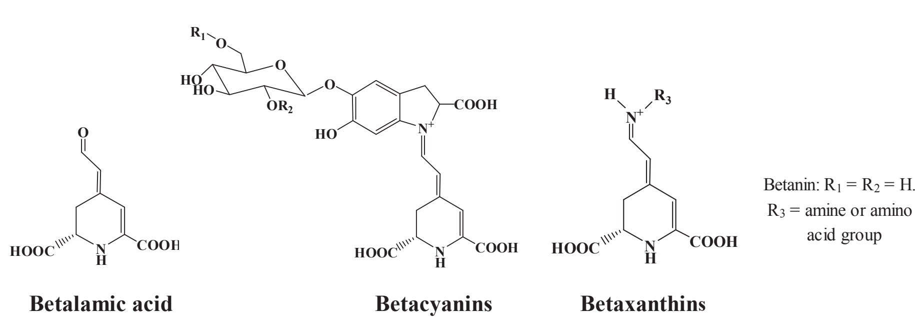

Fig. 1. Chemical structures of betalamic acid, betacyanins and betaxanthins.