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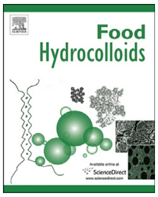

Impact of melting point of palm oil on mechanical and water barrier properties of gelatin-palm oil emulsion film

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A B S T R A C T

In order to explore the effects of oil's melting points on film performance, a gelatin emulsion film with different degrees (8°, 18°, 24°, 33° and 44°, referring to different melting points) of commercial palm oils were fabricated and its properties were investigated. A variety of size distribution ($d_{4,3}$ and $d_{3,2}$) of oil droplets in film-forming emulsion was observed, with 24° oil being the smallest and most regular. As expected, the addition of palm oil obviously decreased film strength but increased elongation, depending upon oil degree. Among them, more importantly, strength of the film containing 24° palm oil was close to that of the pure gelatin film, coexisting with a satisfied elongation value. Furthermore, reducing water vapor permeability was observed in all emulsion films, wherein film containing 24° oil was lowest. Increasing degrees of palm oil led to an increase in film's opacity and a variation of film color, as well as thermal stability. The aforementioned different effects were consistent with microstructure of the emulsion films, where the film with 24° oil droplets was smoother and compact. In conclusion, the melting point of palm oil had a remarkable influence on properties of the resulting emulsion film, mainly being attributed to the differences in size and related state of oil droplets in emulsion during film processing.

1. Introduction

Developing edible packaging, mainly based on a broad spectrum of biopolymers, e.g., polysaccharides and proteins (Chen, Ye, Li, & Wang, 2013; Feng et al., 2014), are receiving increasing interest in food processing because of its biodegradability, biocompatibility and particular use (Fabra, Jiménez, Atarés, Talens, & Chiralt, 2009; Sobral, Menegalli, Hubinger, & Roques, 2001). Different aspects of the raw material for edible packaging must be taken into account when selecting polymer, such as functional properties including solubility, viscosity, and barrier, optical and mechanical properties, health safety, accessibility and price (Matsakidou, Biliaderis, & Kiosseoglou, 2013). Gelatin is obtained from partly acid or alkaline hydrolysis of collagen deriving from a variety of inexpensive animal origins (such as bone, skin and connective tissue) with formula weights from 3 to 200 kDa relying on the crude material used and the extraction process (Lacroix & Cooksey, 2005). Gelatin is a colorless and scentless mixture of protein and polypeptide, which has come into widespread use in the manufacture of sugar coated tablet, hard/soft capsules in pharmaceutical industries (Choi & Regenstein, 2000). Together with good emulsifying properties and the high water binding capacity (Wang, Liu, Ye, Wang, & Li, 2015), gelatin has also been widely utilized in many kinds of food, e.g. candy, sausage, ice cream and instant noodles. On the other hand, gelatin, due to its excellent gelling properties and film-forming ability, is more studied as a prospective candidate for the development of edible film and packaging (Bae, Darby, Kimmel, Park, & Whiteside, 2009). For instance, gelatin-based film prepared from porcine and bovine skin were reported, with low puncture deformation and high puncture strength (Sobral et al., 2001). The capabilities of gelatin-based film vary with the source of gelatin, the types and contents of plasticizer, processing method, etc (Vanin, Sobral, Menegalli, Carvalho, & Habitante, 2005). However, poor barrier property toward water vapor is still, in some ways, restricting the use of various gelatin film, because of the

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hydrophilicity of gelatin and hydrophilic plasticizer, required to make film flexible (Hoque, Benjakul, & Prodpran, 2011).

To minimize the disadvantage of poor barrier property to moisture, the gelatin-based film is incorporated with hydrophobic substances (e.g. oils, fats, fatty acids and waxes) (Feng et al., 2013; Matsakidou et al., 2013; Soazo, Pérez, Rubiolo, & Verdini, 2013; Tongnuanchan, Benjakul, & Prodpran, 2013). Composite structure can be prepared by means of either dual-layer or emulsion: the former process is that the lipid forms the second layer over the gelatin layer and the latter is dispersing the lipid in the gelatin matrix. There is more advantages of emulsified film over the dual-layer ones, such as the simpler preparation techniques and better product performance (Gallo, Debeaufort, Callegarin, & Voilley, 2000). The properties of emulsified film rest with type and quantity of lipids, compatibility and microstructural heterogeneity of components (gelatin and lipid), as well as the more specific preparation techniques (Fabra, Pérez-Masiá, Talens, & Chiralt, 2011). As for the material, it has been reported that a wide range of vegetable oils (e.g. soy oil, corn oil, sunflower oil, rapeseed oil, olive oil) are easily available in that they are renewable, nontoxic and nonvolatile. Furthermore, they contain multifarious unsaturated fatty acids and their incorporation to edible film or coatings may bring various positive health benefits (Ma et al., 2012).

Nevertheless, high price and low yield can constrain many kinds of oil from being applied to emulsion film. It is important to find cheap and abundant lipids, so as to pave a way in improving water-blocking performance of gelatin-based film. Palm oil may be a satisfactory candidate, whose output is maximum comparing with that of all the other edible vegetable oils in the world (Mba, Dumont, & Ngadi, 2015). The oil is extracted from the ripe fruits of oil palm trees, mainly planted in the Southeast Asia, e.g. Malaysia, Indonesia, Thailand. Palm oil is reddish in color and high in saturated fatty acids content, which has widespread use in common food products, e.g. margarines, pastries and fried instant noodles (Edem, 2002). Serving as hydrophobic substances to enhance the water vapor barrier ability of gelatin-based film, palm oil has been studied recently (Tongnuanchan, Benjakul, Prodpran, & Nilsuwan, 2015). They found that the palm oil addition could improve water vapor barrier property of gelatin film. Also, the physical, structural and thermal properties of emulsion film were directly affected by levels of palm oil incorporated. In fact, palm oils are always fractionated to change its physicochemical properties and classified for various applications, by means of redistributing the fatty acids chains with different filtration and selective crystallization methods (Kellens, Gibon, Hendrix, & De Greyt, 2007). On the market, there is a classification system of palm oil according to their differences in melting point, expressed as ° (degree), etc. For example, 8° palm oil indicates the palm oil's melting temperature is approximately 8 °C.

Giving the differences in properties of emulsion films added with a variety of oils and fats owing to variation of fatty acid's chain length and melting points (Fabra et al., 2011; Muscat, Adhikari, McKnight, Guo, & Adhikari, 2013; Seyedi, Koocheki, Mohebbi, & Zahedi, 2015), the addition of different melting points of palm oil into films would make film performances varied, either beneficial or deleterious for a portion or all of parameters. Through delicate comparison of their differences in enhancing film, it would be found that the addition of palm oil with optimized melting point is a practical and valuable way to improve edible film's performances in conventional conditions, such as room temperature (under this temperature, plant oil exists in melted state and easily exudates on film surface, while pre-melted solid fat and wax present a quick crystallization which disturbs film structure, contributing to an inferior performance of emulsion film). So far, with the best of our knowledge, limited information on the aforementioned hypothesis is available in the published literatures. So, the aim of this paper was to study the effects of different melting points of palm oil on the gelatin-plam oil emulsion films' properties, including mechanical strength, water vapor permeability, and morphology, microstructure as well as thermal stability. The obtained results confirmed the rationale of a close relation of melting point and film performance. With the optimization of melting point, a satisfy enhancing with palm oil emulsion for gelatin film was achieved; providing an enough strength equal to conventional film, and coexisting a desirable water resistance.

2. Materials and methods

2.1. Materials

Gelatin (medical grade, gel strength 240 g Bloom) and glycerol (minimum purity 99%) were purchased from Aladdin Industrial Corporation (Shanghai, China). All the palm oils with different degrees (8°, 18°, 24°, 33° and 44°), were kindly donated by Longwit Cereal & Oil Co., Ltd (Tianjin, China). Degrees (°) is a commercial term referring to melting point for palm oil and the degree value of the palm oils used was detected using the slip point method (ISO 6321-2002). Tween-80 was obtained from Beijing Solarbio Science & Technology Co., Ltd. All reagents used for determination of fatty acids composition of palm oil were GC grade and obtained from Sigma–Aldrich (Shanghai, China). Other commercial chemicals are of analytical grade and water is ultra-pure.

2.2. Determination of fatty acids composition

Fatty acids composition of palm oils is determined using GC–MS with a situ methylation according to the published method (Sánchez-Salcedo, Sendra, Carbonell-Barrachina, Martínez, & Hernández, 2016; Trigueros & Sendra, 2015). The detection was conducted with a Model 7890 gas chromatography unit (Agilent Technologies, USA) equipped with a flame ionization detector (FID) and a DB-23 capillary column (30 m length, 0.25 mm internal diameter, 0.25 lm film).

2.3. Preparation and characterization of film-forming emulsions

The film-forming aqueous dispersions contained 3.9 wt% gelatin and glycerol as plasticizer, with a glycerol mass ratio of 33 wt% (based on the protein). The obtained solution was heated at 60 °C for 30 min with stirring every 10 min. To prepare emulsion film, the palm oils of 8°, 18°, 24°, 33°, 44° (preheated at 60 °C until completely melted) with Tween-80 as an emulsifier (Rezvani, Schleining, Sümen, & Taherian, 2013; Soazo et al., 2013) in a 1:4 emulsifier/oil weight ratio were added into the solution previously obtained, respectively, at the level of 36 wt% (based on gelatin content). Then, the mixtures were homogenized at 12,000 rpm for 3 min using a homogenizer (IKA T25 digital ultra-turrax, Staufen, Germany). Finally, the obtained emulsions were degasified for 25 min at 0.09 MPa with a vacuum pump (water circulating pump SHZ-III, Zhixin, China). The ambient temperature throughout the whole experiment was about 25 °C.

2.3.1. Oil droplet size distribution in film-forming emulsions

The lipid particle size of each emulsion was determined using a BT-9300S laser light scattering particles distribution instrument (Bettersize Co., Ltd, China). The volume-weighted mean ($d_{4,3}$) and the surface-weighted mean ($d_{3,2}$) particle diameter of the oil droplets were obtained by the formulas (1) and (2) below (Jiménez, Fabra, Talens, & Chiralt, 2010); n_i is the amount of droplets of a specific size range and d_i is the droplet diameter correspondingly.