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Exploring the properties of modified fish gelatin films incorporated with different fatty acid sucrose esters

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ABSTRACT

Incorporation of hydrophobic materials with surfactant properties such as fatty acid sucrose esters (FASEs) into edible protein films may reduce their drawbacks of having poor water barrier for wider application. In this study, two types of FASEs, PASE and SASE were studied as glycerol substitute in gelatin films at different concentrations (25, 50, 75 and 100%). Interesting to note that high amounts of the FASEs reduced the films' water vapor permeability and solubility but increased opacity. Moisture sorption isotherm revealed that at water activity above 0.75, FASEs improved the water barrier. Meanwhile, tensile strength and Young's modulus were enhanced whereas percentage elongation was decreased. SEM displayed a rougher surface morphology of Gel-PASE and Gel-SASE which was a consequence of disrupted polymeric structure in the film matrix. ATR-FTIR proved the conformational changes in those films as portrayed by the band shifts at 3286–3297 cm^{-1}, 2919–2927 cm^{-1} and 1036–1079 cm^{-1}.

1. Introduction

Packaging is an integral part of food processing that can extend the shelf life and improve the quality of food products. The most often used food packages are those made of non-biodegradable, synthetic materials. However, edible films as packaging materials made from natural polymers are currently gained much interest due to their biodegradability, renewability, consumability and non-toxic properties (Shit & Shah, 2014). In preparation of edible films, plasticizers are always added to improve their handling properties. Commonly used plasticizers are polyols which are very hydrophilic and may contribute to poor water barrier properties of films. Thus, the use of hydrophobic plasticizers has been proposed to not only improve water barrier but also mechanical properties of the films.

Recently, the fatty acid sucrose esters (FASEs) have received a wave of popularity as potential substitutes to hydrophilic plasticizers. FASEs are products of esterification of sucrose to the fatty acids that exhibit non-ionic surfactant characteristics. Since the sugar sucrose consists of eight hydroxyl groups, it can be esterified to 1–8 fatty acids, resulting in formation of FASEs ranging from monoesters to octaesters (Chansanroj & Betz, 2010). FASEs are considered as Generally Recognized as Safe (GRAS) substances, which are organic, tasteless, odourless, biodegradable and non-toxic (Szűts, Budai-Szűcs, Soica, Dehelean, & Szabó-Révész, 2010). Due to these characteristics, FASEs are widely used as additives in many food, cosmetic and pharmaceutical products as they can commercially produced from renewable, low cost and abundant natural resources.

Among the important roles of FASEs in food application is to decontaminate food pathogens and their contact surfaces (Soli et al., 2010). FASEs can also be used as coating agents for various types of fruits to prevent discoloration and chilling injury during storage (Yamauchi, Tokuhara, Ohyama, & Shigyo, 2008). Despite these scientific findings, FASEs have been studied as edible film components aiming for wider application. These edible films include methylcellulose-glucomannan-pectin blend (Chambi & Grosso, 2011), arabinoxylan (Phan The et al., 2002), pullulan (Diab, Biliaderis, Gerasopoulos, & Sfakiotakis, 2001), hydroxypropyl methylcellulose (Villalobos, Hernández-Muñoz, & Chiralt, 2006) and gelatin (Jongjareonrak, Benjakul, Visessanguan, & Tanaka, 2006) based films.

According to European Food Safety Authority (2010), the usage of FASEs as additives in various types of foods and beverages is authorized via Maximum Permitted Levels (MPLs) which are reported varying from 1 to 20 g/kg. However, as for their uses in surface treatments for foods (usually fruits), FASEs are classified under *quantum satis*, where the

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maximum levels are not specified but instead the amounts should not exceed the necessary levels to achieve the intended purposes. In addition, this authority also reported that the FASEs would not exhibit safety concern for human consumption, provided that the overall exposure is within the Acceptable Daily Intake (ADI) of 40 mg/kg bw/day.

To date, studies on the use of FASEs as substitutes to hydrophilic plasticizers are still limited. An early study reported by Jongjareonrak et al. (2006) on the effects of fatty acid sucrose esters on gelatin based films plasticized with 50% glycerol has inspired and motivated us to find more cost effective film formulations but with good performance. It is widely reported that gelatin films usually exhibit good gas barrier and mechanical properties but have poor barrier against water vapor which could limit their applications as packaging materials (Hammann & Schmid, 2014). It is believed that addition of more hydrophobic materials like FASEs into the gelatin films may reduce this drawback.

Current work is aimed at fabricating gelatin films from Tilapia fish (*Oreochromis niloticus*) skins using a lower glycerol concentration of 15% (w/w of gelatin polymer) and elucidating the effects of incorporating different fatty acid sucrose esters on the water barrier, light barrier, mechanical, and morphological properties of the films. The selection of this Tilapia fish skin gelatin as film former is based on the fact that it is considered as low market by-products of food processing and potential environmental pollutants. Moreover, gelatin derived from marine species including fish is preferable over the mammalian gelatin due to the outbreak of bovine spongiform encephalopathy (BSE) disease as well as more acceptable among vegetarians, Muslims and Jews (Norziah, Siti Rashima, & Mehraj, 2015).

2. Materials and methods

2.1. Experimental materials

Tilapia fish (*Oreochromis niloticus*) skins were supplied by a local fish filleting processing company in Perak, Malaysia and brought to the laboratory under chilled condition in ice. Sodium hydroxide (≥99%) and sulphuric acid (95–97%) used for gelatin extraction were bought from QRëc (Selangor, Malaysia) while citric acid (100%) was from Fisher Scientific (Loughborough, UK). Glycerol (≥99%) used as plasticizer in film preparation was purchased from Fischer Scientific (Loughborough, UK). Palmitic acid sucrose ester (P-1570) and stearic acid sucrose ester (S-1570) were kindly provided by Juhalim Biotech Sdn Bhd, Selangor, Malaysia. Glutaraldehyde (25%) and ethanol (99.7%) were purchased from QRëc (Selangor, Malaysia). Sodium cacodylate (98%) and osmium tetraoxide (4%) were purchased from Acros Organics (Geel, Belgium) and phosphorus pentoxide (≥97%) was obtained from HmbG Chemicals (Illinois, USA). All other chemicals used are of analytical grade.

2.2. Production of gelatin

Fish skins were thawed slightly to remove residual meat, fat and scales followed by rinsing under running water and cut into small pieces (50 mm × 30 mm) prior to gelatin extraction following the method described by Grossman and Bergman (1992) with slight modifications. Fish skins were pre-treated by soaking initially in 0.2% sodium hydroxide solution (40 min, 25 ± 2 °C), then in 2% sulfuric acid solution (40 min, 25 ± 2 °C), and lastly in 1% citric acid solution (40 min, 25 ± 2 °C) with washings with tap water in between the soaking steps. Finally, the mixture was adjusted to pH 7 by washing several times under running water before subjecting to hot water extraction (8 h, 45 ± 2 °C) and filtered to remove the skin residues. The filtrate collected was dried in an air-circulated oven (500, Memmert, Germany) for about 18 h at 50 ± 2 °C and ground to 30 mesh size. Fish gelatin powder obtained had a measured protein content of 91.5 ± 0.2% using the Kjedahl method (AOAC, 2005) with a nitrogen conversion factor of 5.5.

Table 1
Film formulations.

Films	Ratio (Glycerol: PASE or SASE)	Weight (g)		
		Glycerol	PASE	SASE
Gel-G15	100:0	0.38	–	–
Films with PASE addition (Gel-PASE)				
Gel-PASE25	75:25	0.28	0.10	–
Gel-PASE50	50:50	0.19	0.19	–
Gel-PASE75	25:75	0.10	0.28	–
Gel-PASE100	0:100	0.00	0.38	–
Films with SASE addition (Gel-SASE)				
Gel-SASE25	75:25	0.28	–	0.10
Gel-SASE50	50:50	0.19	–	0.19
Gel-SASE75	25:75	0.10	–	0.28
Gel-SASE100	0:100	0.00	–	0.38

Gel-Gelatin; G-Glycerol; PASE-Palmitic acid sucrose ester; SASE-Stearic acid sucrose ester. Weight is expressed based on 100 g of film forming solution.

2.3. Preparation of glycerol plasticized gelatin films with addition of fatty acid sucrose esters (FASEs)

Gelatin (2.5%, w/w) was dissolved in water under continuous stirring for 15 min at 50 ± 2 °C. Glycerol as the plasticizer was then added into the solution at a concentration of 15%, based on weight of gelatin. The film mixture was further mixed under constant stirring for another 30 min at 70 ± 2 °C, followed by homogenizing at 13,000 rpm for another 15 min. This control gelatin film was noted as Gel-G15. To study the effects of adding fatty acid sucrose esters (FASEs) on gelatin films, two types of FASEs which are palmitic acid sucrose ester (PASE) and stearic acid sucrose ester (SASE) were added into the film mixture by substituting the glycerol at different levels (25, 50, 75 and 100%). The formulations of the gelatin emulsion films for Gel-PASE and Gel-SASE are given in Table 1. The FASEs were dissolved and mixed thoroughly with a homogenizer (Ultra Turrax T25 Basic, IKA Labortechnik, Malaysia) at 13,000 rpm for 15 min at 70 ± 2 °C before adding into the film mixture solution. For all film forming solutions, after cooling to room temperature at 25 ± 2 °C, 85 g of the solutions were poured onto Perspex plates (160 mm × 160 mm) and left to dry at room temperature for about 48 h. The dried films were peeled from the casting plates and conditioned in a chamber, at 25 ± 2 °C and 55 ± 3% relative humidity using saturated sodium bromide solution for at least 48 h prior to analyses for their water barrier, light barrier, mechanical, and morphological properties. Each film formulation was prepared in triplicates and was compared with control Gel-G15 film. Subsequently, the effects of adding fatty acid sucrose esters as glycerol substitutes, on the film properties were evaluated based on their different concentrations and sizes (carbon chain lengths).

2.4. Characterization

2.4.1. Film thickness measurement

Film thickness was measured using a hand-held micrometer (Mitutoyo, Tokyo, Japan) to the nearest 0.001 mm at twelve different locations and the average value was obtained.

2.4.2. Determination of mechanical properties

Films of each formulation were uniformly cut into strips (150 mm × 25 mm), and tensile strength (TS), Young's modulus (E) and percentage elongation (%E) were measured and calculated according to ASTM D882 method using texture analyzer (TA.XT plus, Stable Micro Systems, Surrey, UK). The film strips were clamped between tensile grips with an initial grip separation and test speed set at 100 mm and 0.8 mm s^{-1}, respectively using a load cell of 30 kg. For each formulation, nine film strips were cut and average of nine measurements was taken to calculate the TS, E and%E.