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Effects of bio-nanocomposite films from tilapia and squid skin gelatins incorporated with ethanolic extract from coconut husk on storage stability of mackerel meat powder

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ABSTRACT

Chemical, physical and sensory changes of mackerel meat powder covered with tilapia and squid skin gelatin films and nanocomposite films incorporated without and with ethanolic extract from coconut husk (EECH) in comparison with that covered with polyethylene (PE) film and the control (without covering) during storage of 30 days at 28–30 °C were investigated. The powder covered with nanocomposite film incorporated with EECH at 0.4% (w/w) (SGF-Na-EECH) generally had the lower moisture content than those covered with other gelatin films ($P < 0.05$). The lower peroxide value (PV), thiobarbituric acid reactive substances (TBARS), total volatile base (TVB) and pH were observed for SGF-Na-EECH sample than PE sample and the control ($P < 0.05$). Based on SPME-GC–MS analysis, SGF-Na-EECH sample contained the lower volatile lipid oxidation products. Higher overall likeness score was observed for SGF-Na-EECH sample on day 30 of storage. Thus, nanocomposite film incorporated with both EECH and nanoclay could be an alternative to synthetic commercial film to maintain the quality and extend the shelf-life of mackerel meat powder.

1. Introduction

Food packaging has been used to facilitate marketing and to protect food products from harmful environmental factors by providing a barrier to mass transfer and mechanical protection (Abreu, Losada, Maroto, & Cruz, 2011). Several food items susceptible to lipid oxidation could be stored for an extended time by the appropriate packaging (Artharn, Prodpran, & Benjakul, 2009). The use of proper packaging technology to minimise quality losses and assure the safety of foods has gained increasing attention (Jongjareonrak, Benjakul, Visessanguan, & Tanaka, 2008). However, packaging materials currently used for food packaging are mostly derived from petroleum by-products, contributing significantly to environmental and ecological problems (Gomez-Guillen et al., 2009). Moreover, there is an increasing concern about the potential risk that plasticiser such as phthalate used in plastics can leach out into foodstuffs. To overcome these problems, biodegradable, non-toxic, natural and renewable sources have

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been searched to replace the synthetic plastic packaging (Tharanathan, 2003; Weng & Wu, 2014).

Gelatin, the partially hydrolysed form of collagen, is mainly extracted from bovine bones, hides and porcine skin generated as byproducts during animal slaughtering and processing (Karim & Bhat, 2009). Fish gelatin has been paid increasing attention as the alternative of land animal gelatin due to religious constraints of mammalian counterpart (Benjakul, Kittiphattanabawon, & Regenstein, 2012). Amongst all biopolymers, gelatin is considered as promising biopolymers under the light of its film-forming ability and applicability (Gomez-Guillen et al., 2009). Due to their inherent hydrophilic properties, gelatin films absorb water at elevated relative humidity (RH) conditions. This results in the plasticised film matrices, but weakens barrier and mechanical properties (Martucci & Ruseckaite, 2009). Several technologies have been implemented for improvement of film properties (Gomez-Guillen et al., 2009; Hoque, Benjakul, & Prodpran, 2011; Nunez-Flores, Castro, Lopez-Caballero, Montero, & Gomez-Guillen, 2013).

Nanoclays, both hydrophilic and hydrophobic, were incorporated with gelatin films to improve their mechanical and water vapour barrier properties as well as thermal stability (Farahnaky, Mohammad, Dadfar, & Shahbazi, 2014; Nagarajan, Benjakul, Prodpran, & Songtipya, 2014a). Due to the enhanced polymer-filler interfacial interaction, nanocomposite films showed improved mechanical and barrier properties and thermal stability (Martucci & Ruseckaite, 2010). Montmorillonite nanoclays, such as Cloisite Na^+, with hydrophilic in nature, could be homogeneously dispersed in a hydrophilic polymer matrix to form the new nanocomposite (Bae et al., 2009; Nagarajan, Benjakul, Prodpran, & Songtipya, 2014b). This forces water and gas travelling through the nanocomposite film via an increased 'tortuous path' of the film matrix surrounding the nanoclay, thereby increasing the effective path length for diffusion (Ray & Okamoto, 2003; Rhim, 2007). Nevertheless, nanoclays with small size, large surface area and high reactivity may cause the adverse effects on health such as cytotoxic effect (Baek, Lee, & Choi, 2012) and genotoxic effect (Houtman et al., 2014). Thus, nanoclays have not been yet approved for human consumption.

Coconut husk is the byproduct, which is either burned for energy production or simply disposed (Vazquez-Torres, Ganche-Escamilla, & Cruz-Ramos, 1992). The preparation of ethanolic extract containing phenolic compounds could increase the value of the husk. Plant phenolics have been used to improve the physical properties of gelatin films (Gomez-Estaca, Lopez-de-Dicastillo, Hernandez-Munoz, Catala, & Gavara, 2014; Hoque et al., 2011; Rattaya, Benjakul, & Prodpran, 2009) and to increase bioactivity of resulting films (Nunez-Flores et al., 2013). Recently, ethanolic extract from coconut husk has been shown to form suitable nanocomposites with gelatin and nanoclay, yielding the films with improved water vapour barrier property (Nagarajan, Benjakul, Prodpran, & Songtipya, 2015). The obtained nanocomposites with ethanolic coconut husk extract could therefore serve as the active packaging for shelf-life extension of fishery products rich in polyunsaturated fatty acids.

To the best of our knowledge, there is no information on the uses of nanocomposite films from fish or squid gelatin containing natural extract in fishery products. Thus, the objective of this investigation was to study the quality changes of mackerel meat powder covered with nanocomposite films from tilapia and squid skin gelatins incorporated with ethanolic extract from coconut husk during the storage time of 30 days at 28–30 °C.

2. Materials and methods

2.1. Chemicals

Fish skin gelatin from tilapia (~240 bloom) was purchased from Lapi Gelatine (Empoli, Italy). Montmorillonite (MMT)-nanoclay, Cloisite® Na^+ was obtained from Southern clay products Inc. (Gonzlaes, TX, USA). Sodium hydroxide, hydrogen peroxide (H_2O_2) (30.96% w/v), chloroform, sodium chloride, trichloroacetic acid and glycerol were procured from Merck (Darmstadt, Germany). Cumene hydroperoxide and malondialdehyde were purchased from Sigma (St. Louis, MO, USA). 2-Thiobarbituric acid, 1,1,3,3-tetramethoxypropane, ammonium thiocyanate and ferrous chloride were procured from Fluka (Buchs, Switzerland). Ethanol, hydrochloric acid, boric acid and di-pottasium carbonate were purchased from RCI Labscan (Bangkok, Thailand). All chemicals were of analytical grade.

2.2. Preparation of squid skin and extraction of gelatin

The skin of fresh splendid squid (*Loligo formosana*) was obtained from Sea Wealth Frozen Food Co., Ltd., Songkhla, Thailand and stored in ice using a skin/ice ratio of 1:2 (w/w). The sample was transported to the Department of Food Technology, Prince of Songkla University, Hat Yai, Thailand within 2 h. Upon arrival, the skin was cleaned and washed with iced tap water (0–2 °C). The skin was then cut into small pieces (0.5×0.5 cm^2), placed in polyethylene bags and stored at −20 °C until use. The skin was stored for not more than 2 months. Prepared skin was subjected to gelatin extraction following the method of Nagarajan, Benjakul, Prodpran and Songtipya (2013). Freeze-dried gelatin contained 97.58% protein (dry weight basis) as determined by the Kjeldl method (AOAC, 2000).

2.3. Extraction of ethanolic extract from coconut husk

2.3.1. Collection and preparation of coconut husk

Coconut husk was obtained from a local market in Hat Yai, Songkhla, Thailand. Husk sample was prepared as per the method of Vazquez-Torres et al. (1992) with slight modifications. Husk sample was dried at 60 °C in the cabinet rotary dryer for 16 h and then defibered. Husk sample was then subjected to grinding using a mill (IKA Labortechnik Colloid Mill, Selangor, Malaysia). The prepared sample was then sieved with the aid of sieve shaker (Model EVJ1, Endecotts Ltd., London, UK) using a sieve size of 6 mm (Woven wire sieves, Endecotts Ltd., London, UK). This coarse form was further blended using a blender (Panasonic, Model MX-898N, Berkshire, UK) and finally sieved using a stainless steel sieve of 80 mesh. The coconut husk powder obtained was further dried in a hot air oven (Memmert, Schwabach, Germany) at 105 °C overnight. The obtained powder was placed in a polyethylene bag, sealed and kept at room temperature until use.

2.3.2. Preparation of the ethanolic extract

Coconut husk powder was subjected to extraction according to the method of Santoso, Yoshie-stark and Suzuki (2004) with a slight modification. Ten grams of husk powder were mixed with 250 ml of 80% ethanol (w/v). The mixture was stirred at room temperature (28–30 °C) using a magnetic stirrer (IKA-Werke, Staufen, Germany) for 3 h. The mixture was then centrifuged at $5000 \times g$ for 30 min at room temperature using a RC-5B plus centrifuge (Beckman, JE-AVANTI, Fullerton, CA, USA). The supernatant was filtered using a Whatman No. 1 filter paper (Whatman International, Ltd., Maidstone, England). The filtrate was then evaporated at 40 °C using an Eyela rotary evaporator (Tokyo Rikakikai, Co., Ltd., Tokyo, Japan). To remove the residual ethanol, the extract was purged with nitrogen gas. The extract was then dried using a Scanvac Model Coolsafe 55 freeze dryer (Coolsafe, Lynge, Denmark) to obtain the dry extract. Dried extract was powdered using a mortar and pestle, transferred to an amber bottle and stored in a desiccator until use. The obtained powder was referred to as 'ethanolic extract from coconut husk, EECH'. EECH