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Cold plasma treatments for improvement of the applicability of defatted soybean meal-based edible film in food packaging

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This study investigated the effects of cold plasma treatment (CPT) using various plasma-forming gases on the physical properties of defatted soybean meal (DSM)-based edible film (DSM film). The effect of packaging smoked salmon using a cold plasma (CP)-treated DSM film on the storage stability of salmon was also evaluated. DSM film was formed by casting film-forming solutions (DSM/glycerol at a ratio of 10:3) prepared by high-pressure (172 MPa) homogenization. Among the O_2-, N_2-, air-, He- and Ar-CPTs, Ar-CPT, which increased elongation and lightness of the DSM film, was optimized against tensile strength, elongation, and water vapor permeability using a response surface analysis. Optimal plasma generation time and power were predicted as 15 min and 400 W, respectively. The tensile strength, elongation, and moisture barrier property of DSM film increased by 6.8%, 13.4%, and 24.4%, respectively, after CPT at optimal conditions. The CPT formed DSM film that decomposed easily and retarded lipid oxidation and hardness reduction of smoked salmon during storage at 4 °C. The results from this study suggest the use of CPT for improving the applicability of DSM film in food packaging.
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1. Introduction

Production of edible films from agricultural process-byproducts for food packaging is of interest because of the high-value commercial use of waste streams (Kang & Min, 2010). Edible films from several agricultural process byproducts have been developed (Gómez-Guillén, Giménez, López-Caballero, & Montero, 2011) including apple peel (Sablani et al., 2009), potato peel (Kang & Min, 2010), defatted mustard seed meal (Hendrix, Morra, Lee, & Min, 2012), and defatted soybean meal (Lee & Min, 2014). Currently, studies are focused on the use of whole byproducts to form edible films, as opposed to components extracted or isolated from byproducts, because the use of whole byproducts will reduce the costs associated with manufacturing biopolymer films (Lee & Min, 2014).

DSM is a soybean byproduct that is produced during oil manufacturing. Development of an edible film using DSM (DSM film) was reported by Lee and Min (2013, 2014). However, the physical characteristics of the film, including inferior tensile and moisture barrier properties, may limit its broad application. Therefore, methods to improve the physical properties of DSM films have been of interest. Furthermore, methods that effectively improve DSM films may be beneficial in enhancing the properties of other agricultural byproduct-based films.

Cold plasma (CP) consists of ultraviolet (UV) photons, electrons, positive and negative ions, free radicals, and excited or non-excited molecules and atoms. In combination, these particles can break covalent bonds and initiate various chemical reactions (Kim, Lee, & Min, 2014). CP treatment (CPT) for the modification of surface properties of polymeric materials is advantageous for several reasons, including free of using hazardous solvents, uniformity of treatment, and no generation of thermal damage when in contact with materials (Desmet et al., 2009; Misra et al., 2015; Morent, De Geyter, Desmet, Dubruel, & Leys, 2011). These advantages allow CPT to be used for heat-sensitive biopolymers (Morent et al., 2011). CPT of polymers can be used for surface functionalization, etching, polymer degradation, and cross-linking (Pankaj et al., 2014a). Surface functionalization refers to the formation of functional groups, such as oxygen- and nitrogen-containing groups, on the polymer surface. This reaction occurs when hydrogen atoms on polymer chains form carbon radicals, subsequently causing oxidation or nitration, which forms oxygen- and nitrogen-containing groups, respectively (Inagaki, Narushim, Tuchida, & Miyazaki, 2004; Pankaj et al., 2014a). During CPT, etching can occur by UV radiation and

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bombardment of energetic particles in CP, including electrons, ions, radicals, and excited atoms/molecules on the polymer surface, consequently increasing surface roughness (De Geyter, Morent, Leys, Gengembre, & Payen, 2007; Inagaki et al., 2004). A balance of the reactions induced by CPT may be modified by adjusting the following parameters: type of plasma, determined by the type of gas used for plasma formation (plasma-forming gas), and the CPT conditions to generate plasma (Inagaki et al., 2004). Oxygen (O_2), nitrogen (N_2), air, helium (He), and argon (Ar) are frequently used for modifying the polymer surface (Desmet et al., 2009; Inagaki et al., 2004). O_2 and N_2 are used for surface functionalization and polymer degradation (Inagaki, Narushima, & Lim, 2003; Paynter, 1998; Riccardi et al., 2003), whereas He and Ar cause a minor chemical effect and are used for etching (France & Short, 1997; France & Short, 1998; Hegemann, Brunner, & Oehr, 2003). Thus, understanding the effects of plasma treatment using these different types of gases would be of value to determine the optimal conditions for CPT (Almazán-Almazán et al., 2006).

Much reported research has focused on improving the physical properties of synthetic polymers using CPT (Pandiyaraj et al., 2015; Shahidi, Ghoranneviss, & Wiener, 2015; Song, Oh, Roh, Kim, & Min, 2015; Yan et al., 2015). However, to the best of our knowledge, no studies have reported the effects of CPT on the physical properties of edible biopolymer films prepared from agricultural process byproducts. Thus, the objectives of this study were to (i) evaluate the effects of CPT using various plasma-forming gases on the physical properties of DSM film, (ii) determine the optimal CPT conditions for improving the tensile and moisture barrier properties of the DSM film, and (iii) investigate the effects of packaging smoked salmon with CP-treated DSM film on salmon quality factors including color, lipid oxidation, hardness, and biodegradability, while stored at 4 °C.

2. Materials and methods

2.1. DSM

DSM was supplied by CJ Corporation (Seoul, Korea). Oil was extracted from soybeans (glycine max) by cold pressing, in which approximately 90% of the oil was removed using a mechanical seed crusher. The material produced during the pressing procedure consisted of irregularly shaped flakes (approximately 1 mm thick and 2–20 mm in diameter). The remaining meal was not further treated prior to use as the base material during film preparation. DSM contained proteins, polysaccharides, water, ashes, and lipids of 45.7, 37.4, 10.0, 6.0, and 0.9 g per 100 g meal, respectively. Compositional analysis was conducted by the Korea Food Research Institute (Sungnam, Korea).

2.2. Film preparation

DSM was ground in a blender (HMF-345[E], Hanil Electric Co., Ltd., Seoul, Korea), sieved to yield a fine powder (<250 μm) and exposed to UV light (40 W) for 1 h on a clean bench to reduce initial microbial load. DSM was then mixed with water (10% [w/w]) and the solution was processed using a high shear probe mixer (Model T25 Ultra-Turrax, IKA-Works, Inc., Wilmington, NC, USA) at 5000 rpm for 5 min. The homogenate was treated once using a high-pressure homogenizer (HPH) at 172 MPa (D.O.S. Inc., Siheung, Korea) to de-polymerize and/or deagglomerate DSM particles in the colloid. Glycerol (Samchun chemical, Pyeongtaek, Korea) was used as a plasticizer and was added to the homogenized hydrocolloid (30% [w/w DSM]). Polysorbate-20 (16.7 hydrophilic-lipophilic balance, Ilshinwells Co., Ltd., Seoul, Korea) was utilized as an emulsifier and was mixed with the DSM hydrocolloid (1% [w/w DSM]). The mixture was homogenized at 5000 rpm for 5 min, followed by heating at 90 °C in a water bath for 30 min. After cooling on ice, the colloid was degassed under a vacuum to generate a film-forming solution. Films were cast by pipetting the film-forming solution, as previously described by Lee and Min (2013). Film thickness was 0.18 ± 0.01 mm, which was measured using a micrometer (Model CR-200, Mitutoyo Co., Kawasaki, Japan).

2.3. CPT system

CPT was performed using a low pressure microwave-powered CPT system (SWU-2, Seoul Women's University, Seoul, Korea), as described previously by Kim et al. (2014). Plasma excitation was created using a microwave generator (2.45 GHz) with a variable plasma generation power of 400–900 W. Chamber pressure was reduced to 667 Pa, followed by the introduction of a plasma-forming gas at 1 standard L/min, which was controlled by a mass flow controller (Model 3660, Kojima Instruments Inc., Osaka, Japan). Single film samples (16.1 cm diameter) were treated individually within the treatment chamber. The temperature of the DSM film samples increased from 23.5 ± 0.6 °C to 27.5 ± 0.5 °C ($\Delta T = 4.0$ °C) during CPT, indicating that the plasma treatment was nonthermal.

2.4. Effects of CPT plasma-forming gases on the physical properties of DSM film

O_2 (>99.9%), N_2 (>99.999%), dry air (21% O_2 in N_2), He (>99.999%), and Ar (>99.999%) were utilized to study the effects of various types of plasma-forming gases on DSM film properties including tensile, moisture barrier (water vapor permeability [WVP]), dynamic mechanical thermal, optical, morphological, and ink adhesion properties. For these particular experiments, plasma generation power and treatment time were 650 W and 25 min, respectively. The test sample dimensions for tensile properties, WVP, dynamic mechanical thermal properties, color, opacity, morphology, and ink adhesion were 8 × 50 mm, 51 mm diameter, 8 × 25 mm, 51 mm diameter, 20 × 30 mm, 5 × 5 mm, and 20 × 30 mm, respectively.

2.5. CPT optimization against DSM film physical properties

The effects of plasma generation power and treatment time on tensile strength (TS), percentage elongation at break (%E), and WVP were evaluated to determine the optimal CPT conditions to achieve those properties. The experiment was designed with response surface methods (RSM) using Minitab (ver. 15, Minitab, Inc., State College, PA, USA). A two variable, second-order center composite RSM design was used to show the interactions of the variables on TS, %E, or WVP for 13 runs. Of these, five were for the center point (Nath & Chattopadhyay, 2007). The effects of individual linear, quadratic, and interaction terms were determined using statistical analysis software (SAS).

$$Y = b_0 + b_1X_1 + b_2X_2 + b_{12}X_1X_2 + b_{11}X_1^2 + b_{22}X_2^2 \quad (1)$$

where b_n are the constant regression coefficients, Y is the reduction in TS, %E, or WVP, and X_1 and X_2 are the plasma generation power (400, 473, 650, 828, and 900 W) and treatment time (10, 14, 25, 36, and 40 min), respectively.

Optimal conditions against WVP, TS, and %E were determined using the response optimizer function in Minitab (Kim et al., 2014).

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