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FULL PAPER

Edible film characteristic from yellowfin skin tuna (*Thunnus albacares*) gelatin enriched with cinnamon (*Cinnamomum zeylanicum*) and roselle (*Hibiscus sabdariffa*)

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Abstract

Fish skin gelatin can be developed as a raw material for developing biodegradable packaging such as edible film. The present study was to investigate the feasibility of the combination of fish gelatin and addition of roselle powder (GFRP), cinnamon powder (GFCP) and cinnamon essential oil (GFCEO) in producing edible film and to determine protective effect to food product e.g. bread on the physicochemical properties of the active packaging edible films. All the films were analyzed for their physicochemical properties and its observation as the active packaging for bread. It was observed that the addition of roselle powder, cinnamon powder and cinnamon essential oil gave a better result in edible film properties than control (without addition). Principal Component Analysis (PCA) revealed the distinction among the physicochemical characteristic of GFRP, GFCP and GFCEO. This study also mapped the several physicochemical characteristics of different edible film as packaging treatment.

1. Introduction

Nowadays, the development of biodegradable packaging such as edible film has come into attention since the past few decades as an effort to reduce packaging waste. Among the suitable edible film include several materials such as gelatin, chitosan, carrageenan, waxes, starch, etc. Fish gelatin is a mixture of peptides and proteins produced by the hydrolysis of collagen extract from several tissues such as the skin of fish. It is commonly used in biodegradable films due to its several reason and advantage such as low melting temperature, low oxygen permeability, good film-forming ability and emulsifying properties (Chin *et al.*, 2017).

In Indonesia, tuna is one of the biggest fish commodities. Tuna skin is one of the by-products of canned tuna processing industry, mainly further used to produce fish waste meals and feeds (Hsu, 2010). As fish skins are a major by-product of the fisheries and aquaculture industry. The skin yield is highly variable according to several factors such as species, fish size and processing styles. Conversion of these aquaculture wastes into value-added products such as gelatin that can be applied as biodegradable packaging such as edible film to yield additional income has both economic and waste management benefits for the fish industry (Shyni *et al.*, 2014).

Application of biopolymers such as fish gelatin is a favorable approach to produce active biodegradable packaging. Although fish gelatin has several weakness mechanical properties, this could be overcome by the addition of apices extracts herbs extract and also essential oils (Adilah and Hanani, 2016; Chin *et al.*, 2017; Nilsuwan *et al.*, 2019). Cinnamon and roselle are spice and herbs commonly used in Indonesia (Rahadian *et al.*, 2017; Ningrum *et al.*, 2019). Spices and herbs extract and also essential oil could improve the mechanical properties such as water barrier properties of hydrophilic biopolymers such as gelatin (Wang *et al.*, 2019).

In this study, cinnamon (*Cinnamomum zeylanicum*) powder and essential oil and roselle powder (*Hibiscus sabdariffa*) were selected for incorporation into gelatin blend films to produce gelatin enriched with cinnamon powder (GFCP), cinnamon essential oil (GFCEO) and roselle powder (GFRP). Therefore, this research aimed to study the effect of cinnamon powder, cinnamon

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essential oil and roselle powder in gelatin. The study was also planned to develop active biopolymer film from gelatin enriched with roselle powder (GFRP), cinnamon powder (GFCP) and cinnamon essential oil (GFCEO) and its effect on the quality of bread.

2. Material and methods

2.1 Materials

Yellowfin tuna (*Thunnus albacares*) skin was provided by Fresh Fish, Yogyakarta, Indonesia. The yellowfin tuna skin was parted into abdominal and dorsal skins, the dorsal skin of yellowfin tuna was used in this study. Cinnamon and roselle powder and cinnamon essential were obtained from the local market, Yogyakarta. All chemicals were purchased from Sigma Chemical Co. All reagents used in this study were analytical grade.

2.2 Preparation of gelatin and edible film

Tuna skin was immediately washed, chopped and frozen at until used. The cleaned skin was treated with an acid solution (acetic acid) at ambient temperature for 24 hrs to remove subcutaneous tissue. After the acid treatment, the skin was neutralized water and washed. For hot-water extraction, six volumes (v/w) of distilled water were added and heated at approximately 80°C for several hours. The extracted solution was heated until its concentrated and the filtered solution was dried at for 24 hrs in a cabinet-air dryer at 50°C. Then the dried gelatin was crushed using a waring blender and then the obtained powder was obtained after 40 mesh. Obtained gelatin powder was mixed with cinnamon and roselle powder (1:1 w/w) then characterized further including the physicochemical properties. There were three different formulas of gelatin film: GFCP (Gelatin Film Enriched with Cinnamon Powder), GFRP (Gelatin Film Enriched with Roselle Powder) and GFCEO (Gelatin Film Enriched with Cinnamon Essential Oil). 1% of GFCP, GFRP and GFCEO were each mixed in the solution containing 2% of gelatin and 3% of glycerol. The edible film solution then poured in Petri dish and dried at 50°C for 24 hrs.

2.3 Preparation of coating to bread sample

In order to have a maximum contact between the active packaging compounds enriched with GFCP, GFRP, and GFCEO and bread slices, active coatings were prepared as a model system to better estimation of weight loss of sample to enhance the shelf life (Noshirvani *et al*., 2017). Several parameters such as weight, color, texture, solubility, thickness of active packaging film then were evaluated for 5 days.

2.4 Physicochemical properties of edible film

2.4.1 Color

The color was measured with Chroma Meter (Minolta CR 400, Japan). The instrument was calibrated with the white calibration plate before the measurement. The internal color (International Commission on Illumination L*, a*, b*) of samples were measured. To determine differences in color values to visually perceived differences, the calculation of ΔE was made (Hidayat *et al.*, 2018).

2.4.2 Texture analysis

Texture parameter of gelatin film was analyzed by Texture Analyzer TK1 Lllyod German. *texture analyzer* was set with pre-test speed 0.5 mm/s; *wait time* 0.5 mm/s; *preload/stress* 1,0000 N; *preload/stress speed* 300,00 mm/min.

2.4.3 Solubility

The film's solubility was determined according to the method of Hanani *et al*. (2017). Each film was cut into strips with specific dimensions of 1 × 3 cm2. The strips were weighed followed by immersion in 50 mL of distilled water under constant agitation at 25°C. After filtration, the undissolved film was dried at 110°C to a constant weight. All measurements were done in triplicates. The solubility of a film was determined using the previous method by Chin *et al.* (2017).

2.4.4 Thickness

The film thickness was measured using a mechanical micrometer (Mitutoyo, Japan) with a precision of 0.01 mm. A total of ten measurements were taken along the perimeter of each film

2.4.5 Application of film on bread and its quality assessment

Edible films were prepared for preparing the final basic wrapping formulation. The sliced (10–12 mm thickness) of white bread was cut into squares of 4 cm^2. The bread slices were then wrapped in the control (without addition), GFRP, GFCP and GFCEO. Again, some slices were wrapped in edible film and some were kept unwrapped. The samples were placed on a table and stored at room temperature (Average temperature of 25±2°C and humidity of 75±2%). The quality of all the bread samples was tested with an interval of 0, 1, 2, 3 4 and 5 days. The following quality parameters such as weight change were checked for all the bread samples.

2.4.6 Principal Component Analysis (PCA)

PCA is one type of multivariate analysis where it involves the transformation of a set of variables to a new coordinate system, in which the new axes are the principle components, following the direction of highest variance in the data set. These principal components are orthogonal to one another and also constructed in such a manner that the amount of residual variation decreases with the increasing number of several principal components (Weldegergis *et al.*, 2011). PCA was commonly used to objectively interpret and compare multiple independent volatile compounds that present in the roselle cultivar studied. The purpose of PCA as one of multivariate analysis in order to decrease the number of descriptors associated with the data set while still explaining the maximum amount of variability present in the data. PCA biplots were also constructed using a statistical package Minitab 17 (Minitab Inc, USA).

2.5 Statistical analysis

Analysis of variance (ANOVA) was performed using the general linear model in Minitab 17 Statistical Software (Minitab Inc., State College, PA, USA) and Tukey's multiple comparison tests for the physicochemical parameters (significance level $p < 0.05$). PCA was also performed using Minitab 17 Minitab Inc, USA

3. Results and discussion

3.1 Color

Color is one of very important characteristic in selecting a suitable food packaging because it affects product aesthetics and consumer acceptance. The appearance of several gelatin films is described in Figure 1. Three parameters that were taken into consideration were lightness (L*), green-red (a*) and blue-yellow (b*) color, and were used as the index to show the color difference between the sample GFRP, GFCP, and GFCEO and control (without addition). Film color could be affected by the type, nature and concentration of biopolymer incorporated with essential oil or spice and herbs extract (Ahmad *et al.*, 2015). The results in the present study showed that the addition of roselle powder, cinnamon powder and cinnamon essential oil will influence the color parameter during the storage of the bread (Table 1). Majority of pigments in roselle is from anthocyanin class. During the storage, this can lead to degradation of anthocyanin pigment that will lead to the changing of color parameter (Sinela *et al.*, 2017).

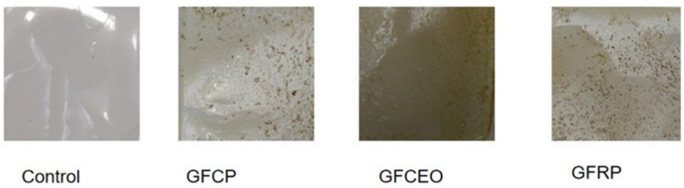

Figure 1. Several edible film formulas e.g. control, GFRP, GFCP, GFCEO

3.2 Texture

Hardness, chewiness and gumminess are evaluated as texture attributes respectively. Table 2 shows

Table 1. The color of edible films for bread packaging with the addition of roselle powder, cinnamon powder and cinnamon essential oil

Treatment	Parameter	Day 0	Day 1	Day 2	Day 3	Day 4	Day 5
Without packaging	L	78.97	78.97	80.58	79.32	76.49	76.27
	a	1.67	1.67	1.87	1.79	2.15	1.89
	b	6.96	6.96	6.15	6.61	6.3	5.82
	ΔE	0	0	10.45	11.04	10.92	10.34
Control	L	72.72	72.72	72.11	71.25	71.39	70.93
	a	3.24	3.24	3.78	4.37	4.03	3.57
	b	22.98	22.98	30.67	26.71	24.08	23.97
	ΔE	0	0	37.96	35.14	32.97	33.22
GFRP	L	57.15	57.15	61.26	58.31	59.43	60.04
	a	6.39	6.39	6.51	5.85	6.13	6.03
	b	26.3	26.3	28.07	23.99	23.65	25.85
	ΔE	0	0	43.15	42.63	41.48	42.34
GFCP	L	61.29	61.29	57.49	61.67	61.97	62.27
	a	7.85	7.85	9.51	7.69	8.02	7.76
	b	29.35	29.35	26.61	28.72	29.68	28.43
	ΔE	0	0	44.9	46.11	43.6	42.45
GFCEO	L	73.31	73.31	71.23	73.17	72.91	70.85
	a	3.05	3.05	2.74	2.75	3.3	2.85
	b	32.37	32.37	25.82	29.52	31.81	28.57
	ΔE	0	0	34.55	36.42	38.49	37.08

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Table 2. The hardness, chewiness and gumminess (N) of edible films for bread packaging with the addition of roselle powder, cinnamon powder and cinnamon essential oil

Sample	Day 0	Day 1	Day 2	Day 3	Day 4	Day 5	Day 0	Day 1	Day 2	Day 3	Day 4	Day 5	Day 0	Day 1	Day 2	Day 3	Day 4	Day 5
	Hardness (N)						Chewiness (N)						Gumminess (N)					
Without Packaging (WP)	52.50	49.44	49.24	57.34	44.86	50.17	41.81	53.24	51.71	36.51	43.84	44.78	46.62	45.98	44.21	41.44	50.02	51.29
Control (C)	47.69	39.69	49.01	59.12	43.59	50.22	37.83	54.14	60.31	31.10	35.78	39.04	42.37	48.61	54.45	35.07	40.45	44.14
GFRP	51.01	36.52	41.03	57.33	34.91	43.65	37.72	46.83	51.56	27.62	29.69	33.36	42.31	41.79	45.93	31.22	33.51	37.74
GFCP	46.64	30.57	36.59	52.53	38.85	47.86	33.73	38.67	45.70	24.09	22.30	25.39	38.01	33.83	40.42	27.26	25.33	28.99
GFCEO	46.67	25.46	30.49	46.10	38.44	47.81	35.13	46.54	55.17	20.22	17.80	21.16	39.96	43.31	42.81	22.91	20.08	24.09

Table 3. The solubility (%) and thickness (mm) of edible films for bread packaging with the addition of roselle powder, cinnamon powder and cinnamon essential oil

Sample	Day 0	Day 1	Day 2	Day 3	Day 4	Day 5	Day 0	Day 1	Day 2	Day 3	Day 4	Day 5
	Solubility (%)						Thickness (mm)					
Without Packaging (WP)	75	75	75	75	75	75	0	0	0	0	0	0
Control (C)	75	75	75	75	75	75	0.20	0.20	0.21	0.20	0.20	0.20
GFRP	71	71	71	71	71	71	0.23	0.23	0.24	0.24	0.24	0.24
GFCP	70	70	70	70	70	70	0.22	0.22	0.23	0.23	0.23	0.23
GFCEO	73	73	73	73	73	73	0.21	0.21	0.21	0.22	0.22	0.22

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hardness, chewiness and gumminess of the gelatin films. The addition of RP, CP and CEO to the films significantly reduced ($p < 0.05$) the hardness, chewiness and gumminess. The texture attributes of the films depend on the intra and intermolecular interactions of the polymer chains in the network. Incorporation of spice and herbs extracts and essential oils as cross-linking agents can lower the texture parameter, as reported before (Lacey *et al*., 2009; Haghighi *et al*., 2019; Xiong *et al*., 2020). The decrease in hardness, chewiness, and gumminess with the addition of RP, CP, and CEO be due to different cross-linking of the polymer during storage. In the present study, the addition of RP, CP and CEO gel disturbed the interaction and alignment among the gelatin chains. This is in agreement with several investigations, in which the properties of gelatin enriched with specific spices and herbs films plasticized with different hydrophilic compounds reported a decrease in texture character of the edible film (Chin *et al.*, 2017; Fakhreddin and Gómez-guillén, 2018).

3.3 Solubility

The solubility of a food packaging film is an important criterion especially for food products with high moisture content. Table 3 shows the solubility of the films. In the present study, the control film showed the highest solubility among all the films and the RP, CP, CEO gel decreased the solubility of the films significantly ($p < 0.05$). This observation was not in agreement with the previous studies, in which the solubility of the films varied proportionally with the moisture content. An explanation for this observation could be fish gelatin is hydrophilic in nature and water molecules were able to permeate the control film which contains only fish gelatin easily, that will influence the solubility of the film (Table 3). The solubility of gelatin films reduced with the addition of different herbs extracts which showed that specific herbs extract might form the strong structure of film network through the high extent of protein–polyphenol interaction (Adilah and Hanani, 2016; Fakhreddin and Gómez-guillén, 2018).

3.4 Thickness

The thickness of each gelatin added with cinnamon and roselle powder and cinnamon essential oil are shown in Table 3. There were no significant differences ($p \geq 0.05$) between the thickness of the control film and the film with the addition of cinnamon and roselle powder and also cinnamon essential oil. The thickness of the film could be influence by the solid content in the film-forming solution that forms the edible film. The peptide bond of fish gelatin could influence the tight network of the film in GFRP GFCP and GFCEO. This means that the components cinnamon and roselle could be distributed in the film without affecting the film thickness (Chin *et al.*, 2017).

3.5 Bread weight loss

The percentage of weight loss in bread samples a function of time, with and without wrapping with the film is shown in Table 4. Wrapping of bread with film reduces the weight loss compared to unwrapped samples; it may be attributed to better WVP (Water Vapor Permeability) of film, which restricted the moisture loss from bread. Addition of cinnamon and roselle powder and also cinnamon essential oil in the film will reduce the weight loss due to protective effect. Addition of selected pomace to the gelatin composite also can reduce the weight loss of bread in the previous study before (Borah *et al.*, 2017).

3.6 PCA

Figure 2A and 2B show the score and loading plot of PCA from several treatments. Each treatment is classified by PCA in a different cluster. There are 5 different clusters (Control, GFRP, GFCP and GFCEO) as described on Figure 2A that. Addition of roselle and cinnamon that contain several essential oil and bioactive compound will also decrease the solubility of edible film. So, the control treatment has the highest solubility other than other groups such as GFRP, GFCP and GFCEO and. For the GFRP and GFCP where there is addition of roselle and cinnamon in powder the edible film. It will give influence directly to the a (redness) value and also the delta E value since anthocyanin as the major natural pigment from roselle can be degraded during storage and also several pigments in cinnamon (Figure 2A).

Table 4. Effect of addition of gelatin edible film with roselle powder, cinnamon powder and cinnamon essential oil wrapping on weight loss of bread during storage and its comparison with unwrapped bread

Sample	Day 0	Day 1	Day 2	Day 3	Day 4	Day 5	Weight Change Percentage (%)
Without Packaging (WP)	1.22	1.05	0.99	0.99	0.96	0.96	21.72
Control	1.42	1.35	1.3	1.3	1.27	1.27	10.38
GFRP	1.3	1.25	1.23	1.23	1.2	1.2	8
GFCP	1.27	1.23	1.22	1.22	1.16	1.16	8.82
GFCEO	1.47	1.44	1.39	1.38	1.35	1.35	8.24

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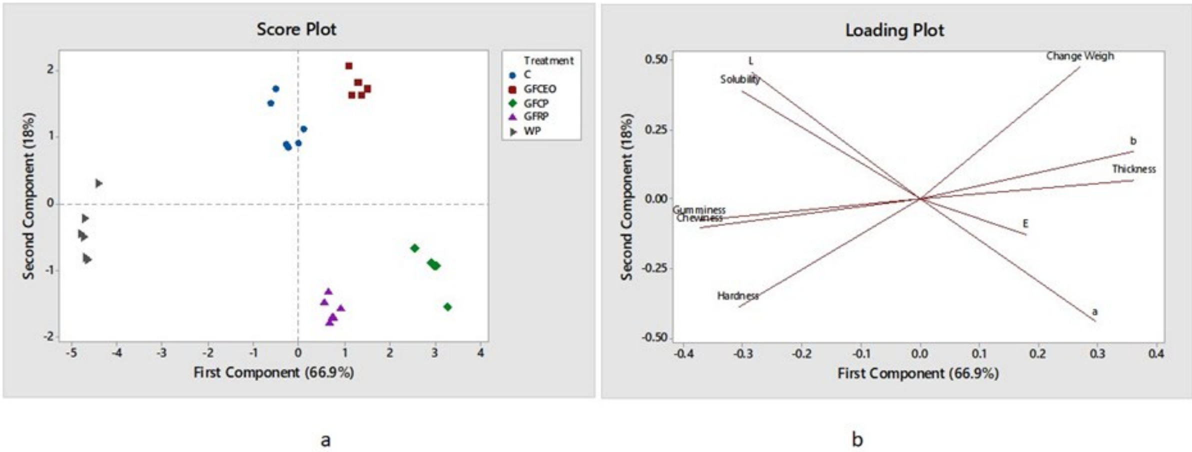

Figure 2. PCA Score Plot (2A) and Loading Plot (2B) of five different treatment (WP, Control, GFRP, GFCP, and GFCEO)

The PCA score plots loadings plot (Figure 2B) shows the first and second principal component of the mean center physicochemical parameters, which together explained more than 84.9% of-of the total variance. The first axis (PC1) described 66.9% of the variance and mainly discriminated a,b, E, thickness, change weight with positives scores and negatives score from hardness, chewiness, gumminess, L, and solubility. The second axis (PC2) described 18% of the variance with positive scores to mainly the parameter such as L (Lightness), b value, solubility, change the weight of bread and thickness, where negative score from hardness, chewiness, gumminess, delta E, and a.

4. Conclusion

Addition of roselle and cinnamon powder and cinnamon essential oil was successfully employed to fabricate the gelatin edible film. The film prepared with the addition of roselle, cinnamon powder and cinnamon essential oil had better physicochemical characterization than the control (without addition). Wrapping of white bread resulted in less weight loss, which could be attributed to the incorporation of active agent in film forming solution. Cinnamon powder, roselle powder and cinnamon essential oil are promising materials to produce edible biopolymer films and coatings. The properties of this gelatin-based film enriched with cinnamon, roselle powder and cinnamon essential oil can be further improved to meet requirements for its commercial applications in the food industry by optimizing film-forming variables. PCA helped to cluster 5 different treatment ((without packaging) WP, (control/ without addition) C, GFRP, GFCP, GFCEO) based on their physicochemical characterization.

Conflict of interest

The authors declare no conflict of interest in the manuscript.

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