

Mechanical and Antibacterial Properties of Chitosan-PLA Film Containing Cinnamon and Ginger Essential Oil for Milkfish Satay Packaging

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Abstract. In this study the effect of Chitosan-PLA (CH-PLA) film enriching with cinnamon essential oil (CEO) and ginger essential oil (GEO) were analyzed to investigate the mechanical and antibacterial properties. Resulting were improved the tensile strength (TS) effect of CEO and GEO addition to CH-PLA film by decrease about 47.39% for CEO and 66.85% for GEO compared to the film without essential oil. Blending CEO and GEO into CH-PLA film can also increase with increasing essential oil. CEO and GEO oil can increase 2.95 and 2.9 of elongation percentage compared to the film without essential oil on 2%of essential oil. CH-PLA film containing additional of CEO and GEO had no important effect on the CH-PLA film thickness. The film thickness for each essential oil composition varied from 0.10 to 0.13 m, while CH-PLA film containing CEO and GEO decrease total weight loss and bacterial population of milkfish satay compared with uncoated and coated without EO milkfish satay.

Introduction

Milkfish satay is one of a typical local culinary in Banten Province. Is one of the proceed milkfish products by thorn with particular emphasis on the seasoning stabbed in bamboo then steamed or burned [1]. It has valuable nutritional properties as it contains 20% of proteins, 0.72% fats, 28.12 minerals, 0.114% carbohydrate and 75.85% water [2]. Milkfish satay susceptible to be spoiled due to the moisture contents, chemical, and physical factors, however it can impact to the short shelf life. Synthetic antioxidants and preservatives are directly added into food products to extend shelf life and control the microbial growth [3]. Nevertheless, the addition of synthetic preservatives can induce adverse health effects [4]. Because of the repulsion of customer, so the natural preservatives such as antibacterial essential oil has been evaluated to assign its efficiency and offer an opportunity to replace the synthetic preservation in food products.

Essential oil are colorless liquids, and naturally provide in all part of plants including stem, bark, flowers, peel, and seeds [5] [6]. Essential oil are expected can reduce the water vapour permeability of hydrophilic film due to their lipidic nature, they have proved to have some effect on the film properties and provide the antioxidant and antibacterial effects [5]. Incorporate an antibacterial in a packaging materials may be an alternative to control the bacterial growth and to extend the shelf life of food products [7].

Blend films are well known as as thin film with 0.04 to 0.2 mm thickness, it is well known as a sustainable and biodegradable plastic material use in agriculture, forestry, and food packaging [8] [9]. The major constituent of blend film can be classified into three components namely, fat, protein

and polysaccharide e.g chitosan. Chitosan (CH) is a chitin derived polysaccharide with a linear binary heteropolysaccharide composed of (b1,4)-linked 2-acetamido-2-deoxy-glucofuranose (GlcNAc) and 2-amino-2-deoxy-glucofuranose (GlcN). It has received significant attention as a main constituent component of blend film [10] and widely used in medicines, dentistry, food processing chemistry, biotechnology, agriculture and environmental protection [11]. Chitosan (CH) is well known as high versatile material due to its biodegradable, biocompatible, antifungal and antimicrobial activity [12], however chitosan (CH) are typically poor in mechanical properties, fragile, weak barrier properties of film, high moisture sensitivity and water-insoluble material [13][14]. Blending chitosan (CH) and synthetic polymer with good mechanical properties such as poly (ethylene oxide) (PEO), poly (vinyl alcohol) (PVA), and poly (lactic acid) (PLA) can improve the mechanical properties of blend film [15][16].

PLA is biodegradable material which commonly acquired by polycondensation of lactic acid [17]. PLA is classified as Generally recognized as Safe (GRAS), it has been widely used in various application such as sanitary products [18], clinical application [19], and food packaging [20]. Integrate the chitosan (CH) enrich with essential oil blend film and PLA can improve the versatile properties such as mechanical properties and water barrier properties of blend film [21].

Based on the description above, the first part of this work was making chitosan-PLA (CH-PLA) blend film containing cinnamon essential oil (CEO) and ginger essential oil (GEO) as antibacterial agents. Blend film with various essential oil contents were characterized to evaluate the blend film mechanical properties. The resulting edible film applied on milkfish satay was then evaluated the bacterial growth, shelf life and weight loss of milkfish satay.

Experimental

Materials. Chitosan and PLA were obtained from (low molecular weight grade, DD 75–85%) was purchased from Sigma-Aldrich, anhydrous acetic acid, chloroform, and sorbitol were purchased from Merck (Indonesia), cinnamon and ginger essential oil and milkfish satay were purchased from local market (Banten-Indonesia).

Methods

Chitosan Solution. Chitosan solution was prepared by dissolving 2 g of chitosan into 100 ml of 1% acetic acid. The solution was then stirred carefully for 2 hours

PLA Solution. PLA solution was prepared by dissolving 2 g of PLA in 200 ml solvent of chloroform under stirring at room temperature for 2 hours.

Chitosan-PLA (CH-PLA) blend film enriching with CEO and GEO. Chitosan (7% by volume), PLA (1% by volume), and 1% by volume of sorbitol were blended. Then adding 0.5% to 2% by volume of CEO and GEO. The solutions were blended for 1 hour using the overhead stirrer (350 rpm). The composite films were obtained by casting on flat glasses and dried at 40 °C for 2 days.

Coating Process on Milkfish Satay. Milkfish satay was wrapped by chitosan-PLA blend film and packed in commercial boxes. Finally, they were stored at room temperature, and antibacterial blend film effect was evaluated in 10 days.

Result and Discussion

Mechanical Properties Analysis. Mechanical properties analysis test were performed to obtain the elongation at break (%), tensile strength (Mpa), and blend film thickness due to the CEO and GEO ratio for each essential oil composition.

Table 1. Film thickness for each essential oil composition

CH-PLA (%v/v)	Essential oil (%)	Film thickness (mm)
7:1	0	0.10
7:1	0.5 CEO	0.11
7:1	1 CEO	0.11
7:1	1.5 CEO	0.12
7:1	2 CEO	0.13
7:1	0.5 GEO	0.11
7:1	1 GEO	0.11
7:1	1.5 GEO	0.12
7:1	2 GEO	0.13

Film thickness was measured by using micrometer at three different location. Based on table 1, the film thickness for each essential oil composition varied from 0.10 to 0.13 mm. The addition of essential oil components give no significant effect to the blend film thickness [22]. Table 1 shown that additional of CEO and GEO had no important effect on the CH-PLA film thickness.

Table 2. Mechanical properties of CH-PLA blend film incorporated with CEO and GEO

CH/PLA (%v/v)	Essential oil (%)	Tensile Strength (Mpa)	Elongasi (%)
7:01	0	29.8	45.6
7:01	0.5 CEO	22.8	47.8
7:01	1.0 CEO	21.8	48.6
7:01	1.5 CEO	20.8	48.9
7:01	2.0 CEO	20.2	49.2
7:01	0.5 GEO	19.8	46.8
7:01	1.0 GEO	18.9	47.2
7:01	1.5 GEO	18.2	48.2
7:01	2.0 GEO	17.9	48.2

Table 2 shows a representative of film tensile strength and elongation. The influence of 2% of essential oil in CH-PLA film are obviously decreasing the tensile strength. Tensile strength (TS) decrease about 47.39% for CEO and 66.85% for GEO compared to the film without essential oil. Oil incorporation into the film can effects the decrease of film tensile strength properties. CH-PLA film incorporate with essential oil can induce the replacement of stronger polymer-polymer interaction by weaker polymer-oil interaction. This interaction may have caused embrittlement of network structure and hence decreasing film tensile strength [23].

Blending CEO and GEO into CH-PLA film can also effect the film elongation percentage. The addition of higher essential oil concentration into film was observe to be higher elongation percentage. As we can see from the table 2, elongation percentage increase with increasing essential oil. The highest (2%) concentration of CEO and GEO oil can increase 2.95 and 2.9 of elongation percentage compared to the film without essential oil. the increasing of elongation percentage is confirmed by the crystallinity indexes. It is conformational a high crystallinity indexes causes the change of film structure.

Antibacterial Properties. The composition of CEO and GEO was play a very important role in antibacterial activity. The effect of essential oil addition can be shown at table 3 as follows.

Table 3. Antibacterial activity of different essential oil composition

CH/PLA (%v/v)	Essential Oil (%)	Total Weight Loss (%)	pH Value	Bacterial population
0	0	50.1	7.88	NA
7:01	0	29.5	6.23	150
7:01	0.5 CEO	23.1	5.91	63
7:01	1.0 CEO	21.7	5.82	40
7:01	1.5 CEO	20.6	5.8	37
7:01	2.0 CEO	17.6	5.68	15
7:01	0.5 GEO	25.6	6.05	142
7:01	1.0 GEO	21.4	5.89	100
7:01	1.5 GEO	19.9	5.83	19
7:01	2.0 GEO	18.2	5.69	15

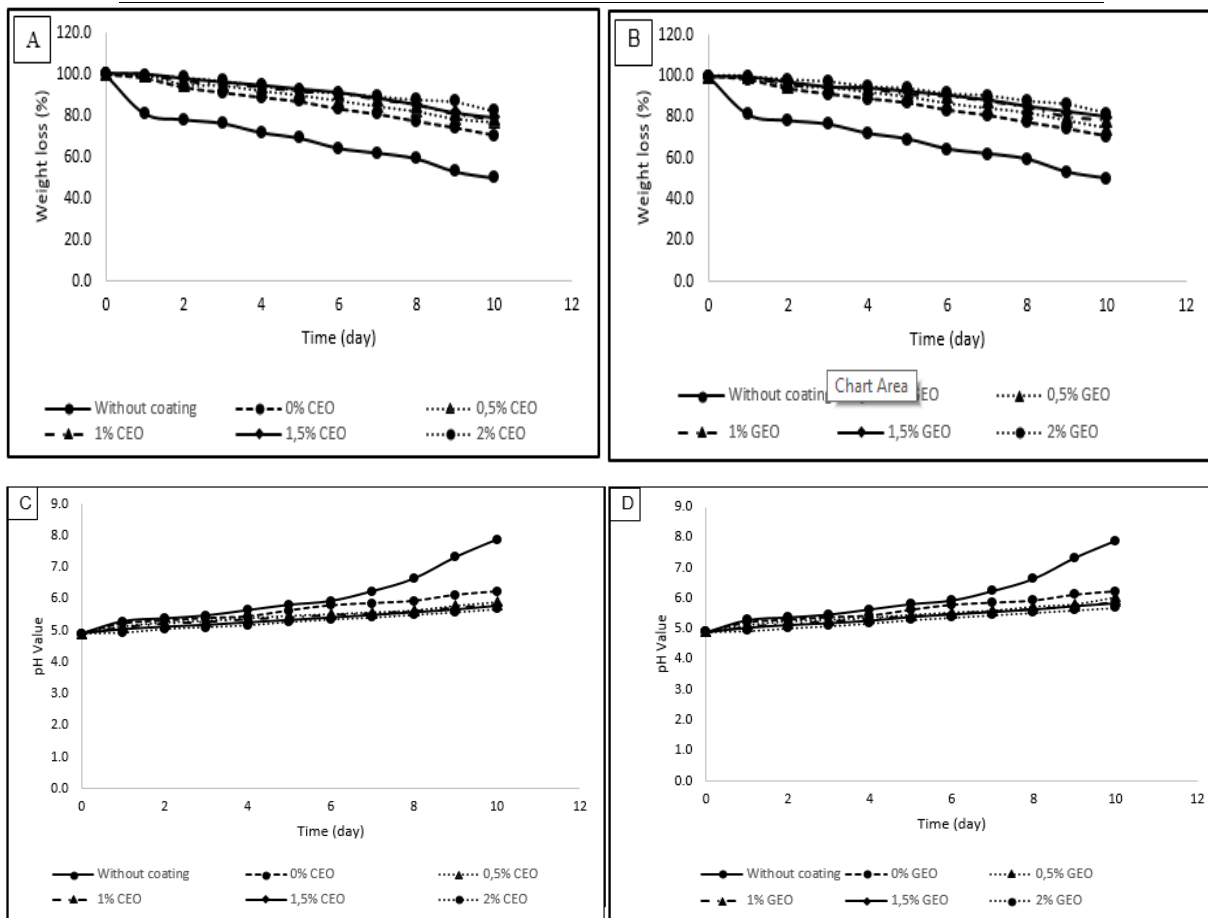


Figure 1. A. Weight loss on different CEO content, B. Weight loss on different GEO content C. pH Value on different CEO content, D. pH Value on different GEO content

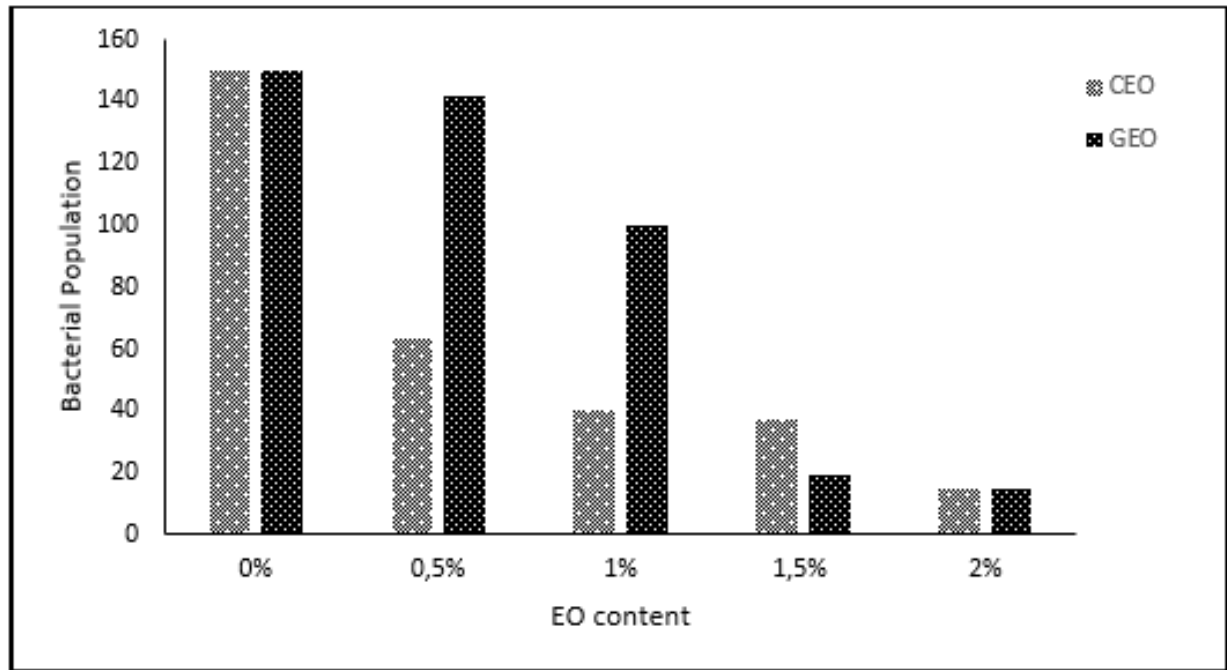


Figure 2. Bacterial population

Table 4. Bacteria performance after treatment

Composition (%)	0	0.5	1	1.5	2
CEO					
GEO					

Six milkfish satay sample per treatment were analyzed to evaluate the weight loss. The samples were weight at the beginning and at the end of each storage periods. The result was then present as the weight loss (%).

Fig. 1 shows the weight loss of coated and uncoated milkfish satay with CH-PLA film. Milkfish satay without CH-PLA coating had statistically higher mass loss compared with CH-PLA coating. The uncoated milkfish satay had 50.07% of weight loss at 10 days of storage. In different composition of essential oil for 1 to 10 days storage at room temperature. Generally, the test samples lost the weight, mainly due to the water diffusion [24]. Weight loss decrease as lipid content increase, it indicates that hydrophobic compound have attempted to improve the moisture barrier. In our study, an increase of essential oil content from 0 to 2% decreasing weight loss, which might the lipid content in coating film leads good moisture barrier performance to gas and water vapor. It also can reduce the respiration and water losses [25].

The effect of essential oil content on pH value indicate in fig. 1C and 1D. As clearly present in this figure, pH value of the milkfish satay were influenced by essential oil concentration and storage day. pH value were increase due to the reduction of essential oil content.

In the non-coated and non-essential sample, each pH value of milkfish satay before and after treatment was increase from 4.8 to 7.88 and 6.23 after 10 days storage. The pH value for coating milkfish satay with 0.5% and 2%CEO are increase from 5.68 to 5.91, while milkfish satay coating with 2% and 0.5%CEO are increase from 5.69 to 6.05 respectively after 10 days storage. The semi permeable properties of essential oil contents can formed the surface of milkfish satay and modified the internal atmosphere such as CO₂ and O₂ concentration. This perform can retarding milkfish satay rottenness.

The application of coating milkfish satay with CH-PLA enriching with CEO and GEO was effective to control the bacterial population (table 3). Milkfish satay with CH-PLA coating enriching essential oil had statistically less bacterial population compared with CH-PLA coating after 9 days storage. [26] reported that food packaging using edible coating based on chitosan effectively decreased the populations of different bacteria and fungi.

Conclusion

The film coating based on CH-PLA enriching CEO and GEO were influence the mechanical and antibacterial properties of film. CEO and GEO can be used to improve film properties. CEO and GEO were effective to control the bacterial population, and provide mechanical properties and product weight loss. This eco-friendly material can be applied in food preservation to extend shelf life and quality of milkfish satay.

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References

- [1] M. T. Ismail and A. Bukhori, "Consumer Perceptions of Sate Bandeng Attributes," in 2nd and 3rd International Conference on Food Security Innovation (ICFSI 2018-2019), 2021, pp. 28–32.
- [2] S. Malle, A. B. Tawali, M. M. Tahir, and M. Bilang, "Nutrient composition of milkfish (*Chanos chanos*, Forskal) from Pangkep, South Sulawesi, Indonesia," *Journal of Nutrition*, vol. 25, no. 1, pp. 155–162, 2019.
- [3] M. Moditsi, A. Lazaridou, T. Moschakis, and C. G. Biliaderis, "Modifying the physical properties of dairy protein films for controlled release of antifungal agents," *Food Hydrocolloids*, vol. 39, pp. 195–203, 2014.
- [4] J. E. De La Torre Torres, F. Gassara, A. P. Kouassi, S. K. Brar, and K. Belkacemi, "Spice use in food: properties and benefits.," *Critical Reviews in Food Science and Nutrition*, vol. 57, no. 6, pp. 1078–1088, 2017.
- [5] L. Atarés and A. Chiralt, "Essential oils as additives in biodegradable films and coatings for active food packaging," *Trends in food science & technology*, vol. 48, pp. 51–62, 2016.
- [6] S. Bhavaniramya, S. Vishnupriya, M. S. Al-Aboody, R. Vijayakumar, and D. Baskaran, "Role of essential oils in food safety: Antimicrobial and antioxidant applications," *Grain & oil science and technology*, vol. 2, no. 2, pp. 49–55, 2019.
- [7] N. Kanani, Rahmayetty, E. Y. Wardhono, and Wardalia, "Preparation and characterization of blend film based on chitosan-poly lactic acid (PLA) composites," in *AIP Conference Proceedings*, 2021, vol. 2370, no. 1, p. 020026.
- [8] A. Sionkowska, "Current research on the blends of natural and synthetic polymers as new biomaterials," *Progress in polymer science*, vol. 36, no. 9, pp. 1254–1276, 2011.

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- [9] L. Abugoch et al., “Shelf-life of fresh blueberries coated with quinoa protein/chitosan/sunflower oil edible film,” *Journal of the Science of Food and Agriculture*, vol. 96, no. 2, pp. 619–626, 2016.
- [10] I. Leceta, P. Guerrero, and K. De La Caba, “Functional properties of chitosan-based films,” *Carbohydrate polymers*, vol. 93, no. 1, pp. 339–346, 2013.
- [11] R. Parhi, “Drug delivery applications of chitin and chitosan: a review,” *Environmental Chemistry Letters*, vol. 18, no. 3, pp. 577–594, 2020.
- [12] A. Alishahi et al., “Chitosan nanoparticle to carry vitamin C through the gastrointestinal tract and induce the non-specific immunity system of rainbow trout (*Oncorhynchus mykiss*),” *Carbohydrate polymers*, vol. 86, no. 1, pp. 142–146, 2011.
- [13] C. Qin, H. Li, Q. Xiao, Y. Liu, J. Zhu, and Y. Du, “Water-solubility of chitosan and its antimicrobial activity,” *Carbohydrate polymers*, vol. 63, no. 3, pp. 367–374, 2006.
- [14] Z. Zakaria et al., “Mechanical properties and morphological characterization of PLA/chitosan/epoxidized natural rubber composites,” *Advances in Materials Science and Engineering*, vol. 2013, 2013.
- [15] M. Vargas, A. Albors, A. Chiralt, and C. González-Martínez, “Water interactions and microstructure of chitosan-methylcellulose composite films as affected by ionic concentration,” *LWT-Food Science and Technology*, vol. 44, no. 10, pp. 2290–2295, 2011.
- [16] J. Xu, J. Zhang, W. Gao, H. Liang, H. Wang, and J. Li, “Preparation of chitosan/PLA blend micro/nanofibers by electrospinning,” *Materials Letters*, vol. 63, no. 8, pp. 658–660, 2009.
- [17] E. Y. Wardhono and N. Kanani, “Development of polylactic acid (PLA) bio-composite films reinforced with bacterial cellulose nanocrystals (BCNC) without any surface modification,” *Journal of Dispersion Science and Technology*, 2019.
- [18] M. Jonoobi, J. Harun, A. P. Mathew, and K. Oksman, “Mechanical properties of cellulose nanofiber (CNF) reinforced polylactic acid (PLA) prepared by twin screw extrusion,” *Composites Science and Technology*, vol. 70, no. 12, pp. 1742–1747, 2010.
- [19] C. E. Tanase and I. Spiridon, “PLA/chitosan/keratin composites for biomedical applications,” *Materials Science and Engineering: C*, vol. 40, pp. 242–247, 2014.
- [20] S. Roy and J.-W. Rhim, “Preparation of bioactive functional poly (lactic acid)/curcumin composite film for food packaging application,” *International Journal of Biological Macromolecules*, vol. 162, pp. 1780–1789, 2020.
- [21] N. E. Suyatma, A. Copinet, L. Tighzert, and V. Coma, “Mechanical and barrier properties of biodegradable films made from chitosan and poly (lactic acid) blends,” *Journal of Polymers and the Environment*, vol. 12, no. 1, pp. 1–6, 2004.
- [22] M. Moradi, H. Tajik, S. M. R. Rohani, and A. Mahmoudian, “Antioxidant and antimicrobial effects of zein edible film impregnated with *Zataria multiflora* Boiss. essential oil and monolaurin,” *LWT-Food Science and Technology*, vol. 72, pp. 37–43, 2016.
- [23] N. Noshirvani et al., “Cinnamon and ginger essential oils to improve antifungal, physical and mechanical properties of chitosan-carboxymethyl cellulose films,” *Food Hydrocolloids*, vol. 70, pp. 36–45, 2017.

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- [24] C. R. Di Franco, V. P. Cyras, J. P. Busalmen, R. A. Ruseckaite, and A. Vázquez, “Degradation of polycaprolactone/starch blends and composites with sisal fibre,” *Polymer Degradation and Stability*, vol. 86, no. 1, pp. 95–103, 2004.
- [25] M. L. Navarro-Tarazaga, A. Massa, and M. B. Pérez-Gago, “Effect of beeswax content on hydroxypropyl methylcellulose-based edible film properties and postharvest quality of coated plums (Cv. Angeleno),” *LWT-Food Science and Technology*, vol. 44, no. 10, pp. 2328–2334, 2011.
- [26] J. Mei, Y. Yuan, Y. Wu, and Y. Li, “Characterization of edible starch–chitosan film and its application in the storage of Mongolian cheese,” *International Journal of Biological Macromolecules*, vol. 57, pp. 17–21, 2013.