

BIOPOLYMER-BASED FILM PREPARATION FOR POTENTIAL SMART FOOD PACKAGING MATERIAL APPLICATION

D. R. Barleany^{1*}, D. K. Sari¹, R. S. D. Lestari¹, Y. Raharja¹, and A. Gunawan²

¹Department of Chemical Engineering, Universitas Sultan Ageng Tirtayasa, Kotabumi, Cilegon 42435, INDONESIA

²Department of Industrial Engineering, Universitas Sultan Ageng Tirtayasa, Kotabumi, Cilegon 42435, INDONESIA

Abstract

Public interest in colorimetric films for food freshness monitoring has increased recently. In addition to extending the shelf life of packaged food products, packaging materials are also required to provide current information about the freshness of the food while ensuring food quality and safety. The current work aims to prepare smart biodegradable films based on biopolymer-containing color indicators to monitor the quality of *Decapterus spp.* The pH-sensing colorimetric film was developed from a chitosan biopolymer modified using polyvinyl alcohol (PVA) and glycerol, as well as methyl red, as an indicator of fish freshness. The effect of using PVA and stirring conditions (temperature and time) on film production was evaluated on its physical appearance, water vapor permeability, and mechanical properties. The results show that the use of PVA can increase the transparency of chitosan films. Incorporating PVA into the film results in brighter and clearer colors compared to films without PVA. The temperature used in the preparation of the film solution has an influence on the mechanical properties and the water vapor permeability. The increasing stirring temperature leads to the enhancement of Young's modulus and the barrier properties against water vapor and moisture, still concurrently impacting a decrease in the film's yield strength and strain. Additionally, the film also exhibits responsiveness to pH during fish spoilage, with a color change that occurs from pink to yellowish. This confirms that the pH-responsive film resulting from this research has great potential to be applied as a real-time indicator of fish freshness during storage.

Keywords: Chitosan, Fish, Food, Packaging, Smart

*Corresponding author: Tel. +62 254 376712

E-mail address: ria.barleany@untirta.ac.id

DOI: 10.22441/ijimeam.v5i3.24030

1. Introduction

Rapid development has been in the food industry, where packaging has become a priority due to various considerations. The general application of packaging aims to preserve food effectively while ensuring high-quality products with a longer shelf life [1]. Food packaging primarily creates a barrier between food and the environment to provide mechanical protection, reducing contact between food and other factors that can cause spoilage, such as microorganisms, oxygen, and UV rays, and preventing loss of taste and odor [2]. For this purpose, plastic film has been widely applied for packaging food products. It plays an important role in protecting factors that cause food spoilage from the environment [3]. However, the large-scale use of plastic film for food and beverage packaging has become the main cause of plastic pollution, which is detrimental to the environment [4].

Biodegradable or bioplastic is considered an effective solution to replace conventional petroleum-

based plastic, reducing various health and environmental problems [5]. Bioplastics developed from natural sources such as animals and plants are being investigated as alternative candidates to conventional synthetic plastics due to their biodegradable and sustainable properties [6]. Natural biopolymers, which include polysaccharides such as alginate, chitin, chitosan, and cellulose, as well as proteins such as gelatin, have received much attention as an alternative material for food packaging [7-9].

Chitosan-based biofilm is a promising candidate for food packaging. It possesses excellent film-forming capacity and has been frequently used as a matrix for composite materials or as a blend with other suitable polymers to obtain the desired properties [7,8]. A study has blended chitosan with polyvinyl alcohol (PVA) and produced films with large elongation at break, low oxygen permeability, high water barrier properties, and good antimicrobial capabilities [9]. PVA is a water-soluble synthetic polymer compatible with chitosan, has good

degradation properties, chemical resistance, film-forming properties, oxygen barrier, and mechanical properties, and has been recognized as safe as a food packaging material [10].

Nowadays, packaging serves as an important protection for food products and helps attract customer interest [11]. The importance of innovation in producing food packaging that suits consumer needs has encouraged the development of new types of sophisticated packaging, including intelligent packaging, active packaging, and smart packaging [2,13]. Smart packaging technology combines intelligent packaging and active packaging, which involves indicators, sensors, and data carriers that can detect changes that occur and monitor the freshness of food products in real time while also being able to extend the shelf life of packaged food due to the active substances contained therein [13,14].

Due to great concerns about food safety and human health, research on biosensors and indicators in smart packaging systems has received increasing attention in the last few years [15]. The smart packaging concept has been tested with promising results for various food products such as chicken fillets [7], fish [16], fruit [17], and milk [18].

Many people consume fish and other marine products owing to their high nutritional content, such as protein, polyunsaturated fatty acids, vitamins, and minerals [16,19]. Unfortunately, high water content and nutritional value can lead to a decrease in the quality of fresh fish during storage due to protein decomposition from microorganism activity [19]. Fish freshness is very important to monitor for consumers, retailers, and industry. Since the techniques used so far are expensive or sometimes require complicated procedures and must be carried out by professionals, so they take a long time [20,21], the development of smart packaging is the right solution for monitoring fish freshness in real-time, which can be done quickly, simply, and at low cost.

Fish that undergo a decomposition process are accompanied by the formation of amines, affecting the pH value [16]. Among the many indicator systems developed recently, the colorimetric pH-responsive indicator has become a hot spot for many researchers [16,22-24]. Several studies have been carried out based on the use of dyes that are sensitive to pH for label or tag film preparation, where a modest change in pH can cause a discernible color change in the film: a colorimetric sensing label based on bromocresol green (BCG) and a sol-gel matrix layer coated onto filter paper was fabricated to monitor the real-time fish freshness [16]; a biodegradable pH indicator film was successfully developed by combining nanofiber-cellulose/chitosan with the addition of methyl red

(MR) dye followed by coating one layer of polylactic acid on the surface to examine beef and fish spoilage [23]; incorporation of bromothymol blue (BTB) dye grafted on a metal-organic framework (MOF) Cr-MIL-101-NH₂ carrier on a film based on cellulose acetate and polyethylene glycol 4000 (PEG 4000) for Grass carp freshness observing under 25°C [24].

Decapterus spp. has been observed to contribute an average of 2% to the total global marine capture fisheries production, and in this context, Indonesia holds the second-largest position after China [25]. Unfortunately, information regarding the appropriate packaging system to ensure the safe consumption of this type of fish has not been found in any reference. Apart from that, the potential for materials with excellent properties from the combination of chitosan with PVA needs to be explored further, especially in terms of its application for food packaging.

In this research, a simple and smart packaging label was developed from chitosan and PVA with MR dye as a pH-sensitive indicator to provide information on fish (*Decapterus spp.*) freshness through color changes during storage at room temperature. The prepared films were also evaluated for their tensile stress and strain, water vapor permeability, and sensitivity to the pH conditions of the storage environment during fish spoilage.

2. Experimental and Procedures

2.1 Materials

Decapterus spp. was purchased from a local market located in Cilegon, Indonesia, food grade of chitosan (degree of deacetylation (DD) = 85%) from PT. Biotech Surindo (Cirebon, Indonesia), fully hydrolyzed PVA, glacial acetic acid 100%, methyl red indicator, glycerol 99.5%, and ethanol 99.9% were supplied by Merck (Darmstadt, Germany).

2.2 Film Preparation

These films were made using conventional casting methods as previously reported by other researchers [26] with modifications. 15 gr of PVA was dissolved in 1000 mL of distilled water and then heated at various temperatures (35, 37, 40, 42, and 45°C) for 60 min, accompanied by continuous stirring until complete gelatinization was achieved. 0.5 mL of glycerol and 10 mL of acetic acid were added to it, then allowed to reach room temperature (30°C)

The indicator solution was made by mixing methyl red with ethanol with a concentration of 15% (v/v), then added to the film solution. 10 g of chitosan was then added and continued with homogenization. Air bubbles were removed from the film solution using a sonicator, then printed using a glass plate measuring

Table 1. Symbols of chitosan/PVA film products

No.	Products	Definition
1	Product 1	Chitosan/PVA film prepared with a stirring temperature of 35°C
2	Product 2	Chitosan/PVA film prepared with a stirring temperature of 37°C
3	Product 3	Chitosan/PVA film prepared with a stirring temperature of 40°C
4	Product 4	Chitosan/PVA film prepared with a stirring temperature of 42°C
5	Product 5	Chitosan/PVA film prepared with a stirring temperature of 45°C

8x12.5 cm, and then dried using an oven at a temperature of 90°C for 18 h. The dried film was then put into a storage cabinet at 30°C and 75% RH before testing. Process variables in making chitosan/PVA films in this study are shown in Table 1.

2.3 Water Vapor Absorption Analysis

Water vapor absorption analysis was carried out by attaching a film label to the top of a glass filled with water at room temperature (30°C) until it covered the entire glass. At every 10-minute interval, measurements of the increase in label weight were carried out until it reached a constant value. Water vapor uptake values were calculated using Equation 1, while the water vapor absorption rate was determined using Equation 2 as follows:

$$\text{Water vapor uptake (\%)} = \frac{(W_t - W_0)}{W_0} \times 100\% \quad (1)$$

where W_t is film weight at time t (g), and W_0 is dry film weight at $t=0$ (g).

$$\text{Water vapor absorption rate (g/min)} = \frac{\Delta W}{\Delta t} \quad (2)$$

where ΔW and Δt are the difference in weight and time respectively.

2.4 pH Sensor Analysis

The film labels were then applied to test their sensitivity to the pH conditions during the decomposition process of the fish during storage for 3 days at room temperature. Fresh fish are sealed in transparent plastic containers with an embedded sensor label. The fish containers were kept at room temperature, and regular daily assessments were conducted. These assessments involved checking the pH levels within the container and monitoring alterations in the sensor label's color.

2.5 Organoleptic Test

Organoleptic analysis of the spoilage process of the fish included odor, texture, and taste. At the same time, the plastic labels were assessed for color changes that occur from acidic to alkaline conditions.

Table 2. Organoleptic assessment score

Score	Odor	Texture	Taste
1	very stink	very flabby	very bad
2	stink	flabby	bad
3	a bit smelly	a bit flabby	not good
4	fresh	chewy	good
5	very fresh	very chewy	very good

The assessment was carried out by the panelists by organoleptic testing standard guide SNI 01-2346-2006 [27], with a scoring guide as listed in Table 2.

2.6 Mechanical Test

The films prepared with different temperatures (35 and 45°C) and stirring times (50, 60, and 70 min) were tested for tensile strength (yield strength and ultimate tensile strength) as well as strain based on the ASTM D882 procedure. Material elasticity was also analyzed through stress (σ) and strain (ϵ) curves. The elastic modulus (Young's modulus) of a material can be determined from the stress and strain values using Equation 3 or what is known as Hooke's Law [28] as follows:

$$E = \frac{\sigma}{\epsilon} \quad (3)$$

where E is the Young's modulus (kN/mm²), σ is stress (kN/mm²), and ϵ is strain (%).

3. Results and Discussion

3.1 Visual Observation of Film Appearance

From Fig. 1, it can be seen that the overall color appearance of the film made from chitosan, both without and with PVA, has a pink color, which comes from methyl red, which was added as an indicator. The visual observation of chitosan and chitosan/PVA loaded with methyl red was also homogenous, thin, and uniform. Strong intermolecular hydrogen bonds between the functional groups (-NH₂ and -OH) of chitosan and (-OH) of PVA is very likely to occur, as evidenced by the appearance of the film, which looks homogeneous, indicating that the two polymer solutions (chitosan and PVA) were perfectly mixed

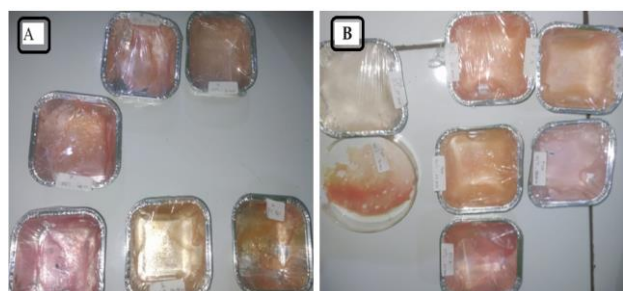


Fig. 1. Chitosan-based film labels (A) without PVA; (B) with PVA addition

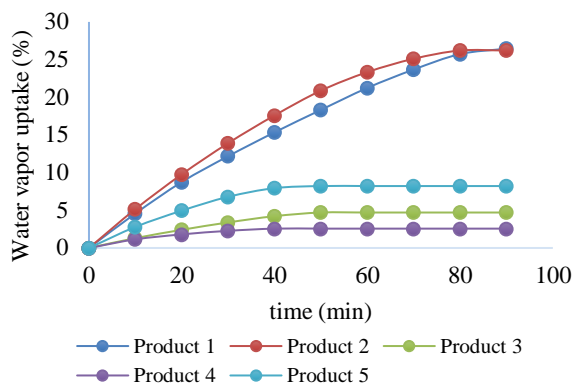


Fig. 2. Water vapor uptake of chitosan/PVA film products

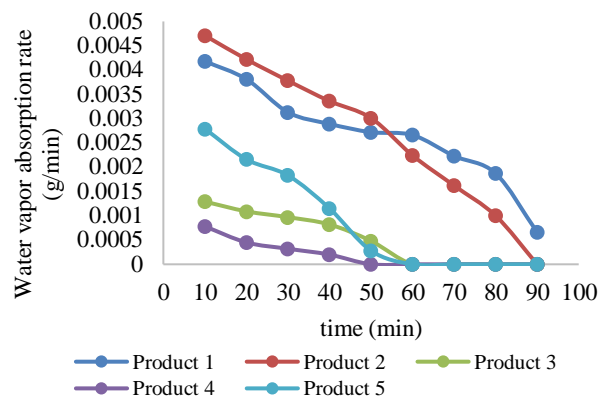


Fig. 3. Water vapor absorption rate of chitosan/PVA films

[26,29].

In this study, all films (chitosan and chitosan/PVA) were fabricated using consistent quantities and ratios of methyl red. The distinguishing factor among these film variants lies in the treatment during the mixing process, incorporating variations in heating temperature (35, 37, 40, 42, and 45°C). Notably, films containing PVA (Fig. 1(B)) exhibit enhanced clarity compared to those without PVA (Fig. 1(A)). This increased transparency can be attributed to the presence of PVA, known for its ability to generate films with heightened transparency and lightness [30].

3.2 Water Vapor Absorption

The barrier ability against water vapor is one of the important properties of films for packaging systems, especially food packaging. The film's ability to absorb water vapor needs to be evaluated to find out how big a role it plays in the exchange of water vapor between the food product and the environment and how it affects its mechanical properties [31]. The amount of water vapor absorbed and the rate of water vapor absorption into the film matrix produced with varying mixing temperatures are shown in Fig. 2 and Fig. 3, respectively.

Fig.2 shows the increase in water vapor uptake as a function of time in all films made with different stirring temperatures. An increase in the amount of uptake water vapor can be seen from time to time, with a decreasing rate until a saturation condition occurs (Fig. 3). This result is by a study that reported the water vapor transmission rate (WVTR) of film from nanofibrillated cellulose prepared with aqueous emulsion of two blocked isocyanates [32].

Products 1 and 2 experience a very small increase in water vapor uptake value and even reach a constant starting at 80 min, while product 3, product 4, and product 5 are constant at 40 min. The largest enhancement in film weight due to water vapor

absorption is observed in product 1, which achieve 26.45%, then followed by product 2, product 5, product 3, and product 4 with increases of 26.20%, 8.22%, 4.70%, and 2.57% respectively.

Fig. 3 discloses that the water vapor and moisture absorption rates for films prepared with different mixing temperatures exhibit the same trend. The water vapor and moisture absorption rate tend to decrease with the increase in time. Film preparation with a higher mixing temperature (product 3, product 4, and product 5) provides a lower amount of water vapor uptake (Fig. 2) and water vapor absorption rate (Fig. 3) compared to treatment at a low temperature (product 1 and product 2). However, it appears that product 2 gives rise to a higher water vapor uptake than product 1, and product 4 produces the lowest water vapor uptake and absorption rate compared to the other four products. The study's examination of how treatment temperature impacts film water vapor absorption did not present a gradual progression of values from the lowest to the highest temperature or vice versa. Nevertheless, observing the patterns depicted in Fig. 2 and 3 indicates that elevating the mixing temperature during the film solution preparation stage is bound to reduce the film's absorption capacity for water vapor. This means it can enhance the film's ability to act as a barrier against water vapor and moisture.

The desired film to be applied to food minimizes moisture exchange between the food and the environment and vice versa to ensure food safety [33]. Due to its being soluble or expanding in water, the polysaccharide group is considered a material unsuitable for packaging applications. Still, the facts about chitosan show something different. Chitosan film for food packaging is known to be resistant to moisture during storage [34].

3.3 pH Observation

As fish spoilage occurs, there is a gradual change

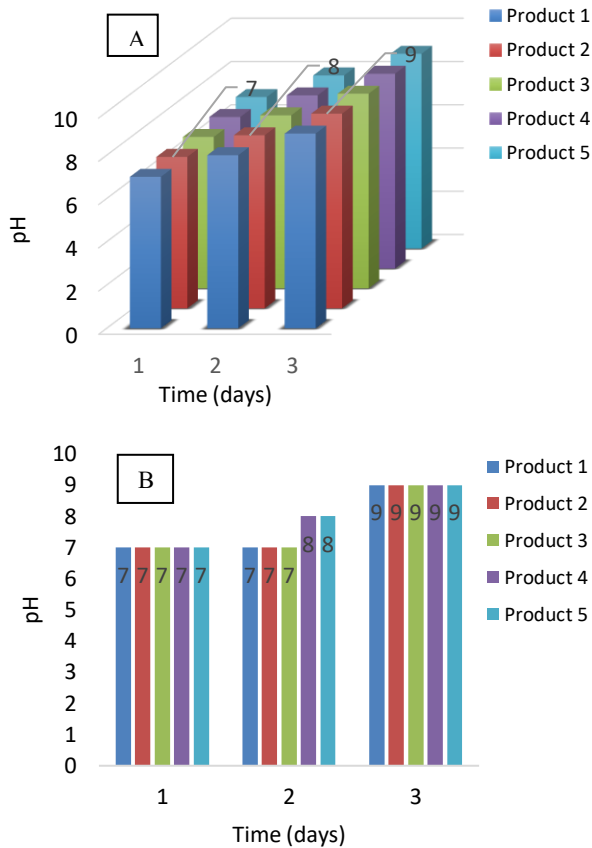


Fig. 4. pH values of fish and labels during spoilage time

in the pH value of the fish and food packaging, which is associated with the accumulation of ammonia, amines, and other nitrogen compounds in the fish, which evaporate into the air, which then causes an unpleasant odor [16,23,35]. In this study, the fish was packaged with a label attached to it and stored at room temperature to observe pH changes that occurred for 3 days. The pH values of the fish and the film label are shown in Fig. 4.

Fig. 4 indicates a gradual increase in the pH value from 7 to 9 from the first day to the third day, and there is no significant difference between the pH measured on the fish and the pH on the film label. This shows that the pH value in the closed packaged air is the same, and the pH of the fish is detected by the film label inside. The increase in pH is a consequence of the amino compound's degradation, which is related to microbial activity, as reported by several studies regarding observations of fish fillet spoilage during storage [23,36].

3.4 Application for Monitoring Fish Quality

The quality of *Decapterus* spp. was observed organoleptically (Fig. 5) from the first to the third day at room temperature.

Fig. 5 shows that the scores for each criterion decreased gradually from day 1 to day 3, which shows

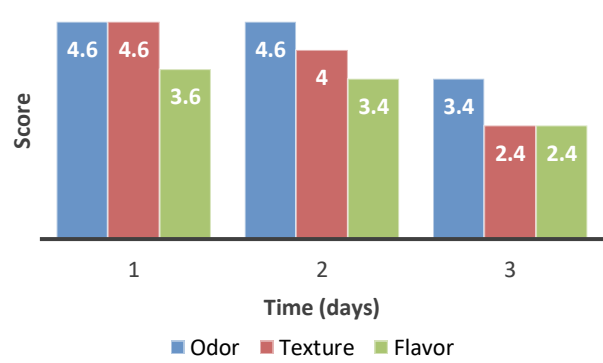


Fig. 5. Organoleptic test results

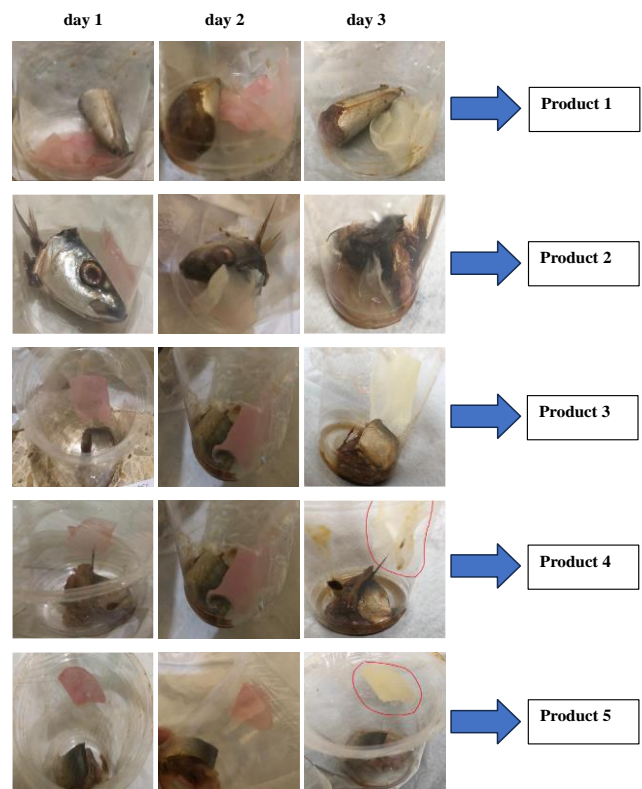


Fig. 6. Color change of the film from chitosan and PVA with methyl red in relation to fish spoilage

a decrease in the subjective level of liking of the panelists. The condition of the fish on day 3 was very bad in terms of smell, texture, and taste. The score displayed is the average of the values given by each panelist.

The color changes of the film during the fish spoilage were also monitored to evaluate the response of the chitosan/PVA sensing label during the fish storage time. As shown in Fig. 6, the film color was initially bright pink. Then, on day two, it faded, and on day 3, it became paler to yellowish. These results were confirmed by a study that monitored fish freshness using a biodegradable indicator from cellulose-nanofiber/chitosan dyed with methyl red and coated with poly (lactic acid) (PLA), which

Table 3. Mechanical properties of the film

Samples		σ_y	σ_u	ϵ (%)	Young's modulus
		(kN/mm ²)	(kN/mm ²)		(kN/mm ²)
T	t				
(°C)	(min)				
35	60	0.027	0.076	68.925	0.039
35	70	0.004	0.019	10.150	0.039
45	50	0.005	0.026	24.850	0.020
45	60	0.021	0.097	49.325	0.043

gradually changed color from reddish to yellow in response to changes in pH from 4-8 [23]. The reddish-pink color on the film label was visible to the naked eye when the fish was still fresh and suitable for consumption. When the freshness of the fish decreased, the film label indicator also showed a color change to a slightly pale pink. In this condition, even though the fish still looked fresh, it provided information that it must be sold or eaten as soon as possible. When the fish spoiled, the color sensor film turned yellow, indicating that the fish was not suitable for consumption and must be thrown away.

3.5 Mechanical properties of the sensing film

In numerous material selection cases, mechanical characteristics are a crucial and obligatory criterion [37,38]. Film's mechanical properties are a very important factor for food packaging products because they are related to their resistance to mechanical stress from consumers themselves during distribution, storage, and treatment [38]. The results of mechanical tests on several random samples of chitosan/PVA sensing film with methyl red are shown in Table 3. The experiment was conducted on four randomly selected samples to assess how variations in temperature and treatment duration during the film solution preparation impacted the mechanical characteristics of the resultant film.

Table 3 contains the mechanical test results of four film samples, which include yield strength (σ_y), ultimate tensile strength (σ_u), and strain. Based on these data, the highest yield strength and strain values were obtained when the film solution was made with a mixing temperature of 35°C and a time of 60 min. In comparison, the highest modulus of elasticity (Young's modulus) value was obtained at a temperature of 45°C and a time of 60 min. Stress and strain curves for modulus of elasticity analysis are shown in Fig. 7.

The Young's modulus values were obtained from the slope of the stress-strain curve in Fig. 7. The process of determining the slope of the area begins from zero up to the yield point, which denotes the juncture where the stress-to-strain curve ceases to maintain linearity. Materials characterized by a high Young's modulus exhibit relatively low elongation, demanding significant stress to induce deformation

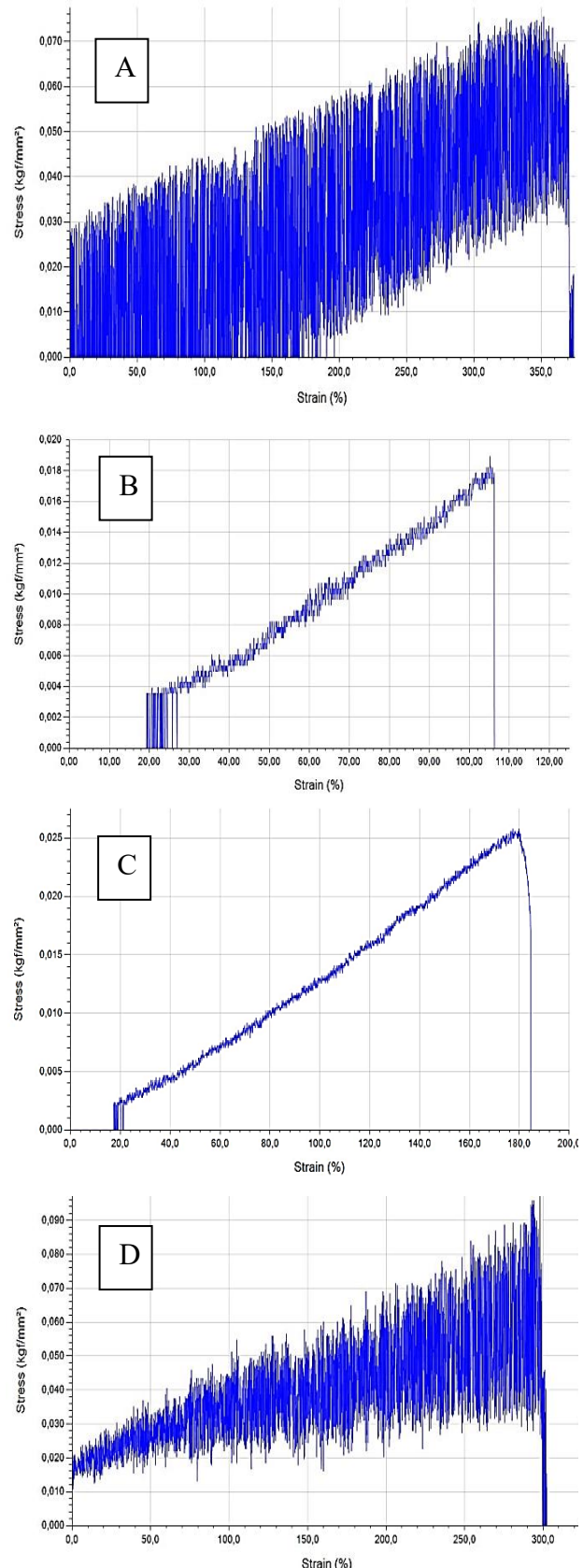


Fig. 7. Modulus of elasticity based on stress and strain curves of varying film preparation treatments(A) $T=35^{\circ}\text{C}$ and $t=60$ min; (B) $T=35^{\circ}\text{C}$ and $t=70$ min; (C) $T=45^{\circ}\text{C}$ and $t=50$ min; (D) $T=45^{\circ}\text{C}$ and $t=60$ min

[28].

In Fig. 7, it's evident that the stress-strain curve's gradient for the product treated at 45°C for 60 minutes of mixing was the most pronounced among the four tested products. This specific film product shows a slope difference of approximately 10.25% compared to products treated at 35°C. Additionally, this study compares the stress-strain curve of the product at 45°C with different stirring durations (50 minutes and 60 minutes). As depicted in Fig. 7C and Fig. 7D, the film subjected to 60 minutes of stirring exhibits a significantly higher slope, twice as steep as that observed with a stirring duration of 50 min.

4. Conclusions

A chitosan/PLA film was successfully developed with methyl red for intelligent food packaging applications. The results reveal that the presence of PVA provides a more transparent color than chitosan film alone. The temperature difference in making the film solution influences the film's mechanical properties and the film's ability to inhibit the transmission of water vapor from food to the environment and vice versa. The higher stirring temperature in the film solution preparation improves Young's modulus and the barrier properties against water vapor and moisture. However, it simultaneously results in a reduction of the film's yield strength and strain. The film produced in this research is also proven to respond to changes in pH through color changes that are visible to the naked eye, so it can be applied as an indicator label for fish freshness in smart packaging systems.

5. Acknowledgements

Acknowledgment conveyed to Universitas Sultan Ageng Tirtayasa for the financial support provided through the 2023 Research Grant (grant no: B/260/UN.43.9/PT.01.03/2023).

References

- [1] Chan, R. B. Y. (2022). Packaging solutions for household food waste in the context of the food/beverage-packaging industry: A comparative review of empirical literature and industry press releases. *Resources, Conservation and Recycling*, 185, 106479.
- [2] Sharma, S., Barkauskaite, S., Jaiswal, A. K., & Jaiswal, S. (2021). Essential oils as additives in active food packaging. *Food Chemistry*, 343, 128403.
- [3] Yan, M. R., Hsieh, S., & Ricacho, N. (2022). Innovative Food Packaging, Food quality and safety, and consumer perspectives. *Processes*, 10(4), 747.
- [4] Gaspar, M. C. & Braga, M. E. M. (2023). Edible films and coatings based on agrifood residues: a new trend in the food packaging research. *Current Opinion in Food Science*, 50, 101006.
- [5] Shaikh, S., Yacoob, M., & Aggarwal, P. (2021). An overview of biodegradable packaging in food industry. *Current Research in Food Science*, 4, 503-520.
- [6] Bhaskar, R., Zo, S. M., Narayanan, K. B., Purohit, S. D., Gupta, M. K., & Han, S. S. (2023). Recent development of protein-based biopolymers in food packaging applications: A review. *Polymer Testing*, 124, 108097.
- [7] Ranjbar, M., Tabrizad, M. H. A., Asadi, G., & Ahari, H. (2023). Investigating the microbial properties of sodium alginate/chitosan edible film containing red beetroot anthocyanin extract for smart packaging in chicken fillet as a pH indicator. *Heliyon*, 9(8), e18879.
- [8] Kongkaoroptham, P., Piroonpan, T., & Pasanphan, W. (2021). Chitosan nanoparticles based on their derivatives as antioxidant and antibacterial additives for active bioplastic packaging. *Carbohydrate Polymers*, 257, 117610.
- [9] Liu, Y., Wang, S., & Lan, W. (2018). Fabrication of antibacterial chitosan-PVA blended film using electrospray technique for food packaging applications. *International Journal of Biological Macromolecules*, 107 (A), 848-854.
- [10] Abdeen, Z. I., El Farargy, A. F., & Negm, N. A. (2018). Nanocomposite framework of chitosan/polyvinyl alcohol/ZnO: Preparation, characterization, swelling and antimicrobial evaluation. *Journal of Molecular Liquids*, 250, 335-343.
- [11] Perera, K. Y., Pradhan, D., Rafferty, A., Jaiswal, A. K., & Jaiswal, S. (2023). A comprehensive review on metal oxide-nanocellulose composites in sustainable active and intelligent food packaging. *Food Chemistry Advances*, 3, 100436.
- [12] Etxabide, A. Kilmartin, P. A., Maté, J. I., & Gómez-Estaca, J. (2022). Characterization of glucose-crosslinked gelatin films reinforced with chitin nanowhiskers for active packaging development. *LWT*, 154, 112833.
- [13] Dirpan, A., Hidayat, S.H., Djalal, M., Ainani, A.F., Yolanda, D.S., Kasmira, Khosuma, M., Solon, G.T., & Ismayanti, N. (2023). Trends over the last 25 years and future research into smart packaging for food: A review. *Future Foods*, 8, 100252.
- [14] Amin, U., Khan, M. K. I., Maan, A. A., Nazir, A., Riaz, S., Khan, M. U., Sultan, M., Munezata,



- P. E. S., & Lorenzo, J. M. (2022). Biodegradable active, intelligent, and smart packaging materials for food applications. *Food Packaging and Shelf Life*, 33, 100903.
- [15] Sobhan, A., Muthukumarappan, K., & Wei, L. (2021). Biosensors and biopolymer-based nanocomposites for smart food packaging: Challenges and opportunities. *Food Packaging and Shelf Life*, 30, 100745.
- [16] Liu, X., Chen, K., Wang, J., Wang, Y., Tang, Y., Gao, X., Zhu, L., Li, X., & Li, J. (2020). An on-package colorimetric sensing label based on a sol-gel matrix for fish freshness monitoring. *Food Chemistry*, 307, 125580.
- [17] Zhang, W., An, H., Sun, X., Du, H., Li, Y., Yang, M., Zhu, Z., & Wen, Y. (2023). Dual-functional smart indicator for direct monitoring fruit freshness. *Food Packaging and Shelf Life*, 40, 101192.
- [18] Pawde, S. V., Chaudhari, S. R., & Mathe, R. S. (2023). Micro perforation based smart label to guide freshness of pasteurized milk packet. *Food Control*, 151, 109783.
- [19] Yumnam, M., Hatiboruah, D., Mishra, R., Sathyaseelan, K., Nath, P., & Mishra, P. (2023). A Smartphone-based optical sensor with polyaniline label for quantitative determination of freshness of freshwater fish fillets. *Sensors and Actuators A: Physical*, 114557.
- [20] Lv, X., Ma, H., Sun, M., Lin, Y., Bai, F., Li, J., & Zhang, B. (2018). A novel bacteriocin DY4-2 produced by *Lactobacillus plantarum* from cutlassfish and its application as bio-preservative for the control of *Pseudomonas fluorescens* in fresh turbot (*Scophthalmus maximus*) fillets. *Food Control*, 89, 22-31.
- [21] Shi, C., Qian, J., Zhu, W., Liu, H., Han, S., & Yang, X. (2019). Nondestructive determination of freshness indicators for tilapia fillets stored at various temperatures by hyperspectral imaging coupled with RBF neural networks. *Food Chemistry*, 275, 497-503.
- [22] Gopika, P. N., Radhakrishnan, P., Baiju, P., & MubeenaS, A. (2023). Fabrication Of Butterfly Pea Flower Anthocyanin-Incorporated Colorimetric Indicator Film Based on Gelatin/Pectin for Monitoring Fish Freshness. *Food Hydrocolloids for Health*, 100159.
- [23] Sobhan, A., Muthukumarappan, K., & Wei, L. (2022). A biopolymer-based pH indicator film for visually monitoring beef and fish spoilage. *Food Bioscience*, 46, 101523.
- [24] Wang, G., He, H., Xu, J., Wang, X., Zhang, T., Huang, S., Li, H., Zhao, P., & Liu, X. (2022). Preparation of fish freshness colorimetric indicator label based on the dye of BTB grafted on MOF carrier. *Sensors and Actuators B: Chemical*, 354, 131230.
- [25] Food and Agriculture Organization of the United Nations. (2022). *The state of world fisheries and aquaculture: Towards blue transformation*. FAO.
- [26] Bhat, V. G., Narasagoudr, S. S., Masti, S. P., Chougale, R. B., Vantamuri, A. B., & Kasai, D. (2022). Development and evaluation of Moringa extract incorporated Chitosan/Guar gum/Poly (vinyl alcohol) active films for food packaging applications. *International Journal of Biological Macromolecules*, 200, 50-60.
- [27] Faisal, M. Gani, A., & Mulana, F. (2019). Preliminary assessment of the utilization of durian peel liquid smoke as a natural preservative for mackerel. *F1000 Research*, 8(240), 1-9.
- [28] Zahroh, F. F., Sugihartono, I., & Safitri, E. D. (2019). Young's modulus calculation of some metals using molecular dynamics method based on the morse potential. computational and experimental. *Research in Materials and Renewable Energy*, 2(1), 19-34.
- [29] Annu, Ali, A., & Ahmed, S. (2021). Eco-friendly natural extract loaded antioxidative chitosan/polyvinyl alcohol based active films for food packaging. *Heliyon*, 7(2), e06550.
- [30] Diop, C. I. K., Beltran, S., Sanz, M-T., Garcia-Tojal, J., & Trigo-lopez, M. (2023). Designing bilayered composite films by direct agar/chitosan and citric acid-crosslinked PVA/agar layer-by-layer casting for packaging applications. *Food Hydrocolloids*, 344, 108987.
- [31] Tavares, K. M., Campos, A. d., Luchesi, B. R., Resende, A. A., Oliveira, J. E. d., & Marconcini, J. M. (2020). Effect of carboxymethyl cellulose concentration on mechanical and water vapor barrier properties of corn starch films. *Carbohydrate Polymers*, 246, 116521.
- [32] Souza, G. d., Belgacem, M. N., Gandini, A., & Carvalho, A. J. F. (2022). Synthesis and characterization of nanofibrillated cellulose films modified with blocked isocyanates in aqueous media and their barrier properties to water vapor and oxygen. *Carbohydrate Polymer Technologies and Applications*, 4, 100249.
- [33] Júnior, L. M., Silva, R. G. d., Vieira, R. P., Alves, R. M. V. (2021). Water vapor sorption and permeability of sustainable alginate/collagen/SiO₂ composite films. *LWT*, 152, 112261.
- [34] Nishida, R., Hirota, N., & Nagai, K. (2023). Anti-cluster behavior and its mechanism during

- water vapor sorption in chitin and chitosan membranes. *Carbohydrate Polymer Technologies and Applications*, 5, 100312.
- [35] Zhuang, S., Hong, H., Zhang, L., Zhang, L., & Luo, Y. (2020). Spoilage-related microbiota in fish and crustaceans during storage: Research progress and future trends. *Comprehensive Reviews in Food Science and Food Safety*, 20(1), 252-288.
- [36] Tan, C., Hu, J., Gao, B., Zhang, B., Li, P., & Shang, N. (2023). Effects of the interaction between *Aeromonas sobria* and *Macrocooccus caseolyticus* on protein degradation of refrigerated sturgeon filets: Novel perspective on fish spoilage. *LWT*, 183, 114908.
- [37] Purnama, M. & Sudjadi, U. (2022). Mechanical study of 9CR-316SS-1MO for nuclear power reactor fuel cladding materials. *International Journal of Innovation in Mechanical Engineering and Advanced Materials*, 4(2), 38-45.
- [38] Martins, V. G., Romani, V. P., Martins, P. C., & Filipini, G. D. S. (2019). Innovative packaging that saves food. In C. M. Galanakis (Ed), *Saving Food: Production, supply chain, food waste and food consumption* (pp. 171-202). Academic Press.