The Synthesis of Edible Film from Tilapia Fish (Oreochromis niloticus) Bones for Ecofriendly Material of Halal Packaging Applications

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Abstract: Packaging materials, especially in food is the most important of healthy aspect. However, today many types of packaging materials used are hazardous to health and environment, then was found alternative halal packaging materials that are safe for health and environment. One of the alternatives is edible film, synthesis from tilapia bone gelatin, it is using sorbitol as plasticizer. Edible film synthesized by dissolving tilapia fish (Oreochromis niloticus) from bone gelatin in distilled water containing sorbitol, then stirred, poured on flat glass, and heated at 50°C. The procedure is performed with a concentration of gelatin (8% and 10% w/v) and sorbitol (25% and 37% v/v). Then was characterized it is mechanical properties include thickness, tensile strength, and elongation to determine eligibility. The optimum condition of the edible film was obtained from 10% concentration of gelatin with 37% sorbitol, with the character of it is mechanical properties value are 0.05 mm thickness, 3.07 MPa tensile strength, and 64.34% elongation.

Keywords: edible film, gelatin, sorbitol, tilapia bone

1. Introduction

Religious belief is one of the factors that influence product purchase decisions (Talib & Johan, 2012). For a Muslim, the halalness of the selected product is one of the considerations. Global halal trends have increased, not only for food products but also in various other products such as clothing, medicines, cosmetics, finance, and even tourism (Talib & Johan, 2012). This increase is evident in the large demand for halal products in the global market, for example in food products whose value is not less than USD600 billion. This happens because besides being consumed by Muslims with a population of more than 1.5 billion people, halal food is also consumed by non-Muslims, because halal food products are cleaner, healthier, and tastier.

The use of the term halal in food is not only related to the raw materials used, but also to the preparation, process, packaging, transportation and storage (Ashadi, 2015). The halal context in the packaging aspect, aims to protect goods from any cross-contamination between halal and non-halal materials. In addition, packaging also functions as protection from all damage to halal products so that they remain in halal status during transportation, distribution, storage until they reach consumers. The second role of packaging is the marketing function. Packaging is basically used in marketing for the purpose of packaging sales, attracting end consumers and strengthening the product image. Packaging is also the most important factor in purchasing decisions for a product (Talib & Johan, 2012).

In general, food product packaging is made of plastic. However, recently there have been a number of efforts to reduce the use of plastics because they are considered dangerous or not environmentally friendly. Therefore, food packaging technology has switched to using materials that are more environmentally friendly and even edible, so as to reduce waste generation from food packaging. The alternative packaging materials that are safe for health and the environment, that material is edible film, synthesis from a gelatin. Gelatin has the properties to form a thin elastic layer, forming a film that is transparent and powerful, has the properties of high digestibility as well as safe for health and safe for the environment because it is easily biodegradable. A common commercial gelatin produced from pork, but it is not halal for muslim. Therefore, an alternative source is needed to obtain halal gelatin, one of which comes from fish bones (Herpandi, Huda, & Adzitey, 2011). In the fish processing process,
bones, scales, and fins are usually waste, even though the amount can reach 30% of the total mass of fish (Zakaria & Bakar, 2015).

Alternative source of gelatin that can be used is from tilapia bone gelatin because the hard fish bones contains 15-17% collagen (Fitriyani, Nuraenah, Lasmi, & Nofreena, 2019). Therefore, more research is needed to synthesis of edible film from tilapia bone gelatin with plasticizer sorbitol. Edible films are elastic, not stiff, protected from damage and can be obtained through the addition of plasticizers (Julianto, Ustadi, & Husni, 2011). Plasticizer is a compound that has a small molecular weight that can be added to the edible film to increase flexibility and mechanical properties of edible film (Kramer, 2009). Plasticizer commonly used in the synthesis of edible film from gelatin is glycerol, sorbitol, triethylene glycol, glucose, and polyethylene glycol (Julianto et al., 2011). The addition of a plasticizer is to obtain the highest value of the extension. This research conducted to determine the optimal conditions edible film of tilapia (O. niloticus) bone gelatin using a plasticizer sorbitol as ecofriendly of halal packaging application.

2. Materials and Methods
2.1 Materials
The raw materials of tilapia bones used in this study were obtained from tilapia fish (O. niloticus) purchased from Bandung local market, West Java, Indonesia. Other materials are hydrochloric acid (HCl 5%), distilled water, and sorbitol obtained from Pudak Scientific, Bandung West Java.

2.2 Experimental Procedures
2.2.1 Gelatin Extraction of Tilapia Bone
The extraction of gelatin from tilapia bone refers to (Deanti et al., 2018) with some modification, includes four stages, that are degreasing, demineralization, extraction, and drying. At first, tilapia bones cleaned from flesh which is still attached to fish bone with cooked for 30 minutes at a temperature of 30-80℃. After fish bone is cooked then separated from the flesh. The size of fishbone was reduced in to the size of 1-2 cm to enlarge the surface area. Bones have been eliminated it’s oil, then demineralization by soaking in a solution of 5% HCl for 48 hours to form ossein. Ossein is washed using distilled water until it has a neutral pH. For further ossein which has had a neutral pH put in a beaker and added with distilled water with ratio of distilled water and ossein are 1: 3 (w/v). After that, ossein heated in a water bath at a temperature of 90℃ for 7 hours before filtered. The filtrate was dried using freeze-drying technic to form gelatin powder.

2.2.2 Analysis of Gelatin
Gelatin is obtained was reacted with a solution of NaOH and CuSO₄ and K₂Cr₂O₇ to prove that the results from freeze-dry is positive gelatin. 0.1 grams gelatin was tested by dissolving distilled water in a test tube. 1 mL NaOH and 2 mL CuSO₄ was added. If solution produced purple color, it was positive solution of gelatin. To further assure that the results obtained are gelatin, then do other evidence by reacting gelatin with K₂Cr₂O₇. 0.1 grams gelatin was tested by dissolving distilled water in a test tube and then added about 3 mL K₂Cr₂O₇. A yellow precipitate that is formed indicates a positive presence of gelatin.

2.2.3 Edible Film Synthesis from Tilapia Bones Gelatin
Edible films made from tilapia bone gelatin with the addition of sorbitol as a plasticizer. The concentration of gelatin is 8% and 10% (w/v) and the concentration of sorbitol used was 25% and 37%. The process of synthesis edible film begins by dissolving sorbitol in water then heating at 70℃, and stirring during the heating process. Gelatin was added until dissolved. Homogeneous solution was poured onto a glass plate. Then drying on an oven at 50℃.

2.2.4 Characterization Edible Film Synthesis from Tilapia (O. niloticus) Bones Gelatin
Edible film thickness measurement uses a micrometers. Edible film thickness individually determined from the average. The unit used is millimeters (mm). Tensile strength test and this extension using Tensilab 5000. The steps of this measurement are the end of samples was clipped by the tensile testing machine and then the length and thickness of the initial samples are recorded. Pressed the start
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button on the computer and then the tool will pull the sample until the sample broke. The tensile strength can be calculated by the following equation:

\[
\tau = \frac{F_{\text{max}}}{A} \tag{1}
\]

Information:
\( \tau \) : tensile strength (MPa)
\( F_{\text{max}} \) : maximum force (N)
\( A \) : surface area (mm\(^2\))

Measurement of the extension is the same way to the measurement of tensile strength. Extension can be calculated by the following equation:

\[
\text{elongation (\%)} = \frac{\text{end length} - \text{initial length (mm)}}{\text{initial length (mm)}} \times 100\% \tag{2}
\]

3. Results and Discussion

Gelatin extraction from tilapia (\( O. \text{niloticus} \)) bones was carried out in several stages. First, the tilapia bones that have been separated from the meat are heated in water with a temperature of 80\(^\circ\)C for 30 minutes. This aims to remove fat from the tilapia fish bones. The choice of temperature at 80\(^\circ\)C is based on the temperature of fat freezing (Rahayu & Fithriyah, 2015). The next stage is demineralization or removal of calcium salts present in tilapia bones. Tilapia bones are immersed in a 5\% HCl solution, the ratio of tilapia bones to HCl is 3:1 (w/v) for 48 hours until ossein or soft bones are formed. The reactions that occur in the demineralization process are as follows:

\[
\text{Ca}_3\text{PO}_4 \text{ (s)} + 6\text{HCl (aq)} \rightarrow 3\text{CaCl}_3 \text{ (aq)} + \text{H}_3\text{PO}_4 \text{ (s)}
\]

The result of the reaction produces dissolved calcium salts and makes tilapia bones soft. The use of HCl as a solvent aims to convert collagen into a form suitable for hydrolysis (Martianingsih & Atmaja, 2010), as well as to break the bonds between carboxyl groups and amide groups in collagen molecules (Siburian, Rochima, Andriani, & Praseptiantangga, 2020)

The next phase is washed by ossein with running water until it has a pH of 5-6, namely the isoelectric pH of the non-collagen component (Siburian et al., 2020). Furthermore, ossein extraction was carried out using distilled water in a water bath with a ratio of ossein and distilled water 1:3 (w/v) for 5 hours at a temperature of 90\(^\circ\)C. This process aims to eat away the hydrogen bonds between molecules that are still not completely decomposed when immersed in HCl. Hydrogen bonds that have completely broken down will produce a water-soluble gel (Martianingsih & Atmaja, 2010). The next step is to separate the gelatin from ossein by filtering it. The gelatin solution that had been filtered was then frozen and frozen for 24 hours. Drying aims to remove water from the gelatin. 158.14 grams of tilapia fish bones can produce 18.35 grams of white color and odorless gelatin powder, so the yield produced is 11.60\%. Gelatin was analyzed with NaOH solution and reacted with CuSO\(_4\). In the second analysis, it was reacted with K\(_2\)Cr\(_2\)O\(_7\) to prove that the result obtained was gelatin.

Optimization of the synthesis edible film from tilapia bone gelatin by using plasticizer sorbitol was done by varying the concentration of gelatin and a sorbitol. The results show at Table 1.

| Mechanic properties | Edible Film |
|---------------------|-------------|
|                      | Gelatin 8% Sorbitol 25% | Gelatin 8% Sorbitol 37% | Gelatin 10% Sorbitol 25% | Gelatin 10% Sorbitol 37% |
| Thickness (mm)       | 0.050        | 0.059        | 0.049        | 0.045          |
| Tensile strength (MPa) | 2.450        | 2.710        | 3.000        | 3.070          |
| Elongation (%)       | 51.66        | 62.66        | 59.33        | 64.34          |

Based on the characterization results (Table 1), the optimum conditions for making edible films from tilapia bone gelatin were obtained in the addition of 10\% gelatin and 37\% sorbitol. The data in Table 1 also shows that in small gelatin formulations, the function of sorbitol can be seen as indicated by its greater elongation value, whereas in larger gelatin formulations, the role of sorbitol is less visible.
because gelatin tends to be more active in hydrogen bonding with other monomers on the edible film, as happened in the polyblend film of breadfruit-chitosan starch (Setiani, Sudiarti, & Rahmidar, 2013).

Table 1 shows that the edible film with a lower gelatin formulation had a higher thickness with the addition of sorbitol, whereas in the formulation with higher gelatin the addition of sorbitol tended to decrease the thickness of the resulting edible film. The increase in sorbitol concentration causes the thickness of the edible film layer to increase. Like water, sorbitol molecules will occupy cavities in the matrix and interact with the polymer (Sudaryati, Mulyani, & Hansyah, 2010). Meanwhile, the mechanical properties of tensile strength experienced an increase in the addition of sorbitol to both lower and higher gelatin formulations.

The increase in sorbitol levels in each formulation caused an increase in the elongation of the resulting edible film. Sorbitol is a plasticizer which prevents the polymer chains in the edible film matrix from forming a very strong bond so that the ability of the edible film to be stretched decreases. Sorbitol is a hydrophilic plasticizer so that it can interact with the edible film matrix. Its ability to interact will prevent the polymer chains from interacting strongly or crystallizing which can cause the film matrix to become stiff. The results Bourtoom (2008) (Wang, Wei, Chang, Sun, & Zhu, 2016) research show that an increase in the concentration of plasticizers (sorbitol) results in an increase in elongation at break. This increase in elongation means that there is an increase in the ability of the edible film to be rolled because the edible film has increased its ability to be stretched.

This research is limited to the mechanical characteristics of the edible film produced, therefore further study is needed to determine the characteristic characteristics of the chemical properties and the ability of edible film as a barrier against the environment such as humidity, water, oil, aroma, and mass transfer, so it can be used in halal packaging technology.

4. Conclusion
Edible films from tilapia (O. niloticus) bone gelatin have been successfully synthesized under optimum conditions of 10% gelatin and 37% sorbitol. The mechanical characteristics of the edible film produced had a thickness of 0.05 mm, tensile strength of 3.07 MPa, and elongation of 64.34%. Further research needs to be carried out to determine its chemical properties and the ability of edible film as a barrier against the environment such as humidity, air, oil, aroma, and mass transfer, so it can be used in halal packaging technology.

Acknowledgement
We acknowledge Examination Laboratory Of High School Textiles University Bandung for the support and cooperation to this research project.

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