Physical Quality of Edible Film from Duck Feet Gelatin Using Various Glycerol Concentrations

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Abstract:
Edible film is a type of packaging that can be consumed, can coat food products, and can be decomposed naturally (biodegradable). This research aims to assess the effect of applying glycerol on the physical quality of edible film from duck's feet gelatin. This research used a completely randomized design (CRD) followed by the Tukey test. The treatment was the use of 10% (T₁), 20% (T₂), 30% (T₃), 40% (T₄), and 50% (T₅) glycerol. The treatments were repeated three times. The findings indicated that the use of glycerol had a significant difference in the yield, thickness, tensile strength, and Young's modulus of edible film (P<0.05), but was not significantly different in the elongation of edible film (P>0.05). The optimal glycerol concentration for edible film in this study was obtained by using 10% glycerol.

Keywords: Physical quality, edible film, duck feet, glycerol.

Introduction
The edible film is a thin sheet that functions as packaging for food products, is easily decomposed naturally (biodegradable), and is environmentally friendly. The mechanism for gel formation in protein-based edible films is that the protein is denatured by heating and endothermic polymerization occurs, followed by surface dehydration. The polymerization process involves disulfide molecules and hydrophobic bonds. The heating treatment causes a three-dimensional protein structure between sulfhydryl and hydrophobic groups, so the unfolded protein chains will approach each other and connect via disulfide and hydrophobic bonds. The protein denaturation process makes the protein chains open, thus allowing the formation of a new matrix network that is more compact and can interact with other components.

The edible film is made from materials that are safe for health so that the packaging can be consumed together with the packaged food. Edible films are made from natural materials, such as polysaccharides, proteins, fats, or a combination of several materials (composites), with or without the addition of plasticizers such as glycerol, sorbitol, sucrose, and others (Cerqueira, et al., 2011).

Gelatin is a hydrolysis product of collagen found in skin, bones, and meat. Gelatin, as one of the ingredients used to make edible films, is transparent, strong, flexible, and able to inhibit the transfer of oxygen and carbon dioxide (Gela, 2016). The benefits of gelatin for food products are as a thickener, stabilizer, emulsifier, binder, adhesive, and edible film packaging. Gelatin is made by hydrolyzing collagen from skin, bones, and animal flesh using acid or alkaline solutions. Animal skins that are widely used in making gelatin are pork skin and cow skin, but according to Islamic beliefs, pork skin is not halal or forbidden for consumption (Abdullah, et al., 2016). The contribution of gelatin from pork skin is 41%, beef bones 30%, cow skin 28.6%, and the rest from fish (Swaidatul, et al., 2016). Another alternative material that is halal to consume to make gelatin is by using by-products from livestock, such as duck feet. Duck feet can be used as raw material for making gelatin because they contain collagen in the skin, bones, and tissue. The collagen protein content in shank skin is more than 80% (Purnomo, 1992). Gelatin made from duck feet has advantages apart from its high collagen protein content, that it is easy to obtain and economically very cheap.
The raw material to produce edible films is made from protein from livestock which has good characteristics and has the potential to be used as a raw material, one of which is livestock feet, by going through a gelatinization process on the collagen protein found in the cattle feet (Miwada, et al., 2015). Protein-based films, like other biopolymers, are limited by a lack of mechanical strength and a poor water vapor barrier due to their hydrophilic nature (Wittaya, 2012). The characteristic of gelatin is that it is brittle or easily broken, so plasticizers need to be added to improve its elastic properties and increase the flexibility of the film.

A plasticizer is added to reduce the stiffness of the polymer and increase the flexibility of the film. Plasticizers prevent film cracking during handling and storage (Vieira, et al., 2011). Glycerol is a plasticizer that is widely used in making edible films. A plasticizer's ability to diminish polymer interactions is proportional to its concentration, type of polymer, and type of plasticizer (molecular configuration, molecular size, number of free hydroxyl groups, and compatibility of the plasticizer with the polymer) (Kokoszka and Lenart, 2007). Using glycerol as a plasticizer is based on the nature of glycerol which is environmentally friendly and non-toxic, therefore it can be beneficial both in industrial processes and in handling it as waste in the environment (Bonnardeaux, 2006).

The good quality of edible film is that it has biodegradable properties (easily degraded or broken down naturally), is easy to consume, and has elastic and flexible properties, with a thin sheet shape or no more than standard film thickness. Edible film yield analysis is conducted to find out the percentage of film products produced and predict the need for raw materials used. The yield value is influenced by the amount of water content that evaporates from the material during the drying process, the greater the value of the edible film can be due to the greater the water content in the edible film. The better the ability of the molecules that make up the material to bind water chemically, the greater the yield value of the material. The interaction between plasticizer and polymer is influenced by the molecular size, configuration, and quantity of plasticizer hydroxide functional groups, as well as the bond with the polymer (Yang and Poulson, 2000).

Several studies have been carried out regarding edible films containing glycerol. Research on making edible film from a mixture of sheepskin gelatin and agar with the addition of 40% glycerol produced a yield of 2.140%, a thickness of 0.097 mm, a tensile strength value of 6.312 MPa, an elongation of 78.130%, and an elasticity value (Young's modulus) of 8.080 MPa. (Fera and Nurkholic, 2018). These results were obtained from a composition of 75% gelatin and 25% agar. The elongation, tensile strength, elasticity and thickness in the edible film research meet the edible film standards which refer to JIS (Japan Industrial Standard). Edible film from split cowhide gelatin with the addition of 40% glycerol and a gelatin weight of 4 grams produces a solubility of 4.677%, a film thickness of 0.11 mm, a tensile strength of 0.99 MPa, and an elongation of 82.66% (Wulandari, 2016). The results of this research also meet edible film standards referring to JIS (Japan Industrial Standard), with a thickness of less than 0.25 mm, tensile strength of more than 0.39 MPa, and elongation of more than 50%, which means very good. Producing edible film using gelatin from duck's feet using glycerol as a plasticizer has not been widely published, therefore this research examines the physical quality of the edible film from duck's feet gelatin using glycerol as a plasticizer.

**Material and Method**

The materials used are gelatin from Cihateup duck (*Anas platyrhynchos javanica*) feet (made by myself), hydrochloric acid and glycerol (Merck, Germany), and distilled water. The tools used were a water bath, drying oven (Julabo, Germany), micrometer screw (Mitutoyo, Japan), and Mesdan Lab Strength Tensolab 5000 version (Italy).

**Edible Films production**

A water bath was used to heat 30 milliliters of distilled water at a temperature of 55°C, gelatin was weighed at 6% (w/v), then dissolved while stirring for 30 minutes, then glycerol with a concentration of 10% (T<sub>1</sub>), 20% (T<sub>2</sub>), 30% (T<sub>3</sub>), 40% (T<sub>4</sub>), and 50% (T<sub>5</sub>) of the gelatin weight was mixed in the gelatin solution. The homogeneous edible film solution was placed into a mold and then dried for 24 hours at 50°C in a drying oven. (Chambi and Grosso, 2006).

**Edible Film Yield**

The yield test was carried out by weighing the weight of the edible film solution and the resulting edible film (dry), then comparing it with the following formula (Fatma, et al., 2015):
Yield (%) = \frac{\text{Edible Film Weight}}{\text{Edible Film Solution Weight}} \times 100\%

**Edible Film Thickness**

The edible film thickness test was measured using a screw micrometer with an accuracy of 0.01 mm. Measurements were taken at five distinct locations on the edible film, and then the average of the film thickness measurement results was taken (Setiani, et al., 2013).

**Tensile strength**

The tensile strength of edible film was determined using the Mesdan Lab Strength tool, Tensolab 5000 version. The tip of the sample was gripped by the test tool, the computer start button was pressed, then the sample was pulled at a speed of 100 millimeters per minute till it broke. Tensile strength is calculated by dividing the greatest stress by the area of the cross-section. To compute the cross-sectional area, multiply the sample's initial length by its initial thickness (Setiani, et al., 2013). Tensile strength is calculated by the formula:

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

Information:
- \( \tau \) = Tensile Strength (MPa)
- \( F_{\text{max}} \) = Maximum Voltage (N)
- \( A \) = Cross Sectional Area (mm\(^2\))

**Elongation**

The film elongation value was measured as the measurement of the film's tensile strength (Setiani, et al., 2013). Film elongation results from the ratio of the gap distance when the film breaks to the initial length of the sample. The following formula was used to calculate elongation:

\[ \text{Elongation (\%)} = \frac{\text{Elongation at break (mm)}}{\text{Initial length of sample}} \times 100\% \]

**Young's Modulus (Elasticity)**

The elasticity of edible film is produced by calculating the tensile strength of the film divided by the elongation of the film (Setiani, et al., 2013). Elasticity is expressed in Mega Pascal (Mpa), with the formula:

\[ \text{Elasticity (Mpa)} = \frac{\text{Tensile Strength}}{\text{Elongation}} \]

**Statistic analysis**

This research was carried out using experimental techniques, utilizing a Completely Randomized Design (CRD), with five treatments, each repeated three times. The data obtained was then checked using variance and then tested using the Tukey test.

**Result and Discussion**

**Effect of Glycerol on Edible Film Yield**

The results of research regarding the application of glycerol as a plasticizer to the physical quality of edible film made from duck feet gelatin with various treatments are presented in Table 1.

<table>
<thead>
<tr>
<th>Yield (%)</th>
<th>Thickness (mm)</th>
<th>Tensile Strength (MPa)</th>
<th>Elongation (%)</th>
<th>Young’s Modulus (MPa)</th>
</tr>
</thead>
</table>

Table 1. Effect of glycerol concentration on the physical quality of the edible film from Duck's feet gelatin
The yield of edible film with various glycerol treatments produced an average of 9.35 - 17.22%. The yield value of the resulting edible film was higher than the yield of the edible film formed from whey Danke and agar with the addition of 25 - 45% glycerol carried out by Fatma, et al. (2015) namely 7.26-7.87%. The Tukey test results showed that the administration of 10% (T1) and 20% (T2) and 30% (T3) glycerol was not significantly different (P>0.05), 20% (T2), 30% (T3), and 40% (T4) were not significantly different, while the 50% treatment (T5) was significantly different (P<0.05) compared to the 10% (T1) and 20% (T2) treatments, but was not significantly different (P>0.05) compared to treatment 30% (T3) and 40% (T4). Increasing the quantity of glycerol as a plasticizer can increase both the total amount of dissolved solids and the concentration of glycerol given. The results are also not significantly different, because the optimum point for adding glycerol has been exceeded, so the effect of glycerol has reached the saturation point, causing it to lose its stability.

The yield percentage increases with increasing glycerol concentration given, this is because the higher the glycerol content, the more dissolved solids and molecules that bind water in the edible film, so that after drying the edible film increases the yield percentage. Glycerol has hydrophilic properties, which means it can bind water, resulting in more water contained in an edible film being bound by glycerol. These results are similar to Fatma, et al., (2015) opinion that the ability of the molecules that make up the material to bind water will cause the yield to be higher. The yield value depends on the water content in the material, the more water content in the edible film, the greater the yield produced. Glycerol can bind water well because the hydroxyl groups can bind water to the polymer chain, so the more glycerol added, the greater the yield of edible film. The interaction between the plasticizer and the polymer is influenced by the molecular size, configuration, and number of plasticizer hydroxide functional groups (Yang and Poulson, 2000). The higher the yield value, the more extract produced. Yield can be used as a benchmark for efficiency in making a product.

**Effect of Glycerol on Edible Film Thickness**

Table 1 shows that the thicknesses of edible film with various glycerol treatments were 0.19 – 0.37 mm. The edible film produced is thicker than the edible film made from chicken bone waste gelatin containing 10 - 40% glycerol carried out by Novian (2018), namely 0.013 - 0.017 millimeters. The thickness of edible film that meets edible film standards referring to JIS (Japan Industrial Standard) is an edible film containing 10 – 20% glycerol, namely 0.19 – 0.21 mm.

The Tukey test results showed that the use of 10% (T1) and 20% (T2) glycerol was not significantly different (P>0.05), as well as the addition of 30% (T3) and 40% (T4) glycerol respectively were not significantly different (P>0.05). The addition of 30% (T3), and 40% (T4) glycerol gave significantly different (P<0.05) higher thickness compared to 10% (T1) and 20% (T2) glycerol, while the addition of 50% glycerol (T5) is significantly different compared to all treatments. This occurs because the quantity of dissolved materials in the mixture increases, affecting the thickness of the edible film. Adding glycerol to edible film enhances its thickness. The large amount of dissolved solids added to edible film will increase its thickness. Using the same mold, the amount of solution poured with different solvent concentrations will produce different thicknesses. In accordance with the opinion of Jacoeb (2014) that the area of the mold, the volume of the solution, and the total amount of solids in the solution all contribute to the thickness of the edible film.

The addition of an increasing glycerol concentration will increase the amount of polymer in the edible film. The increasing thickness is caused by increasing the number of polymers that make up the edible film, the more solids, the thicker the resulting edible film (Tapia-Blacido, et al., 2005). The thickness of the edible film can be adjusted according to the needs of the product to be packaged, by determining the plate or container that will be used as the film mold.
Effect of Glycerol on Tensile Strength of Edible Film

The tensile strength of edible films with various glycerol treatments averages between 0.08 MPa to 2.24 MPa. The tensile strength value of the edible film produced is lower than the tensile strength of edible film made from chicken bone waste gelatin containing 10% - 40% glycerol carried out by Novian (2018), namely 1.45 - 7.73 MPa. The tensile strength of edible film that meets JIS (Japan Industrial Standard) standards is edible film containing 10% (T1) and 20% (T2) glycerol, namely 2.24 MPa and 0.39 MPa. The minimum value of tensile strength based on JIS (Japanese Industrial Standard) (1975) is a minimum of 0.39 MPa.

The Tukey test results indicated that the addition of 10% (T1) and 20% (T2) glycerol had no significant effect (P>0.05), the 20% treatment (T2) up to the 50% treatment (T5) did not significant different (P>0.05), while the 10% (T1) treatment had significantly higher tensile strength values compared to the 30% (T3), 40% (T4), and 50% (T5) treatments. This happens due to the inclusion of glycerol can reduce the forces between molecules so increasing the concentration of glycerol given causes the tensile strength of edible film to decrease. Glycerol is a small hydrophilic molecule that easily fits between molecular chains and forms amide hydrogen bonds with proteins (Qiu-Ping Zhong, et al., 2008). Plasticizers can reduce polymer interactions but depend on the concentration, type of polymer, and type of molecular size. Glycerol has a low molecular weight, so it can reduce the attractiveness of edible films. In accordance with the opinion of Laila (2008) that the attractiveness and efficiency of plasticizers are influenced by their molecular weight, the more the molecular weight of the plasticizer increases, the stronger the attractiveness. The decreasing attractiveness of edible film means that the flexibility of the film increases because the ability of the plasticizer reduces the stiffness of the film.

Tensile strength is a measurement that determines the strength of edible film in protecting packaged food products. A tensile strength value that is too small will reduce its resistance to damage as a food product packaging. The results of the research are similar to edible films made from Jali flour with the addition of a glycerol concentration of 20 - 50% carried out by Anandito (2012), that the tensile strength tends to decrease from a glycerol concentration of 40 - 50% respectively, namely 0.437 - 0.104 MPa. The standard for edible film as packaging for food products is that it has good mechanical properties, but if the tensile strength is too low below the standard, it will reduce its mechanical properties.

Effect of Glycerol on Elongation of Edible Film

The edible film elongation values in Table 1 show that various glycerol treatments produce an average of 72.33 - 95.67%. The edible film elongation value produced from this research is higher than the edible film elongation value made from split cowhide gelatin and soy protein isolate with the addition of 10% - 30% glycerol carried out by Wulandari (2016), namely 42.9 - 88.1%.

The use of glycerol with a concentration of 10 – 50% (T1 – T5) showed no significant difference (P>0.05) in edible film elongation. This can happen because the binding process between glycerol and gelatin molecules is unstable and less effective so it does not have a significant effect. An increase in the hydrophilic protein component of gelatin causes destabilization of the film matrix in the edible film structure (Poeloengasih, 2003). Increasing the stretching of the intermolecular space of the matrix structure in edible film will increase flexibility. Providing the glycerol concentration reaches the optimum point or saturation point, excess glycerol molecules are in a separate phase outside the polymer phase, this causes the intermolecular forces between the chains to decrease, thereby increasing flexibility because the chains move more freely. Increasing the use of plasticizers will increase the edible film elongation value up to a certain concentration (Wiset, et al. 2014).

The results of this research are similar to the production of the edible film made from Jali flour containing 20 - 50% glycerol concentration carried out by Anandito (2012), in that research the elongation value decreased from the addition of 20 - 50% glycerol respectively, namely 41.022 - 13.458%. The edible film elongation value produced meets the standards referring to JIS (Japan Industrial Standard), which is very good because the value is more than 50%.

Effect of Glycerol on Elasticity (Young's Modulus) of Edible Film
The elasticity (Young's modulus) of edible films with various glycerol treatments produces an average of 0.12 – 2.90 MPa. Edible film with a Young's modulus value that meets the standards according to JIS (Japan Industrial Standard), namely edible film using 10% and 20% glycerol (2.90 MPa and 0.42 MPa).

The use of 10% glycerol (T1) was significantly different (P < 0.05) compared to other treatments, while the respective 20 - 50% treatments were not significantly different (P>0.05) to the young edible film modulus value. Using more glycerol causes the binding process between glycerol and gelatin molecules to instability. Glycerol enters the protein chain and forms a protein-plasticizer mixture, disrupting protein-protein interactions, so that the binding of glycerol to gelatin molecules becomes unstable. The properties of glycerol can reduce intermolecular interactions of polymer chains so that the resulting polymer is more elastic.

Conclusion
The use of glycerol produced different edible film qualities in terms of yield quality, thickness, tensile strength, and Young's modulus, but did not produce different elongation qualities. The best use of glycerol in this study was 10%.

Reference