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Optimization of Waste Cooking Oil and Sweet Starter (*Ipomoea batatas*) as Edible Film for Environmentally Friendly Coffee Packaging

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Abstract. Coffee packaging so far has been derived from synthetic polymers. Most synthetic polymers have good oxygen barrier properties, high mechanical resistance, and easy manufacturing processes. However, the weakness of this polymer is that it is difficult to biodegrade so it is not environmentally friendly. Synthetic polymers (plastics) are materials made from petroleum that are difficult to develop and difficult to decompose naturally. One solution to this problem is environmentally friendly packaging, namely edible film. The edible film is a type of plastic that can be consumed and used as a wrapper for a product to reduce the use of plastic. The basic ingredients for making edible films come from starch which belongs to the hydrocolloid group. Indonesia is a country that has starch-producing crops such as corn, rice, and sweet potatoes. This study used sweet potato starch as an ingredient for making edible films. This is because, sweet potato starch has high potential after all in 5 years from 2014-2018 it was the 3rd place as the plant with the highest productivity and had the highest amylose content among other starch-producing plants, namely 38%. The addition of plasticizers or plasticizers is done to improve the rigid and easily torn properties of edible films. One type of plasticizer that is widely used is glycerol. The glycerol used in this study is glycerol from waste cooking oil. Waste cooking oil is the same as coconut oil, so it still contains linoleic acid, stearic acid, and other fatty acids. Waste cooking oil is reacted with a strong base catalyst NaOH in the transesterification reaction to produce glycerol. This research was conducted to utilize waste cooking oil and sweet potato starch as the basic ingredients for making edible films as environmentally friendly coffee packaging.

INTRODUCTION

Packaging is an activity to wrap a product to protect or maintain the product to avoid contamination and damage to the surrounding environment. Plastic is a packaging that is widely used because it has many advantages that are not easily damaged, elastic, transparent, and strong. Plastics come from oil and natural gas (petrochemical polymers) so that they have an adverse impact, where plastics contain hazardous chemicals. In addition, plastic has properties that are difficult to decompose naturally, causing environmental problems [1]. Indonesia is a country that produces plastic waste, including coffee packaging waste. Based on data from the Indonesian Plastic Industry Association (INAPLAS) and the Central Statistics Agency (BPS), plastic waste in Indonesia reaches 64 million tons/year [2]. Therefore, there is a need for alternative packaging products that are safe for human health and easily degraded naturally.

The edible film is packaging in the form of a thin layer and made of materials that can be consumed, safe for health, and environmentally friendly. Edible films can be made from hydrocolloid compounds, fats, and composites. One of the materials that are often used in the manufacture of edible films is starch, which is a hydrocolloid group that is easy to obtain and economical. Starch consists of amylose and amylopectin polymers. Both have different functions, amylose is responsible for the formation of thin films and the formation of strong [3].

One source of starch is tubers such as sweet potatoes. Sweet potato has a fairly high starch content with amylose content ranging from 8.50-37.40% [4]. In addition, according to the Ministry of Agriculture in Indonesia, the productivity of sweet potato for the last 5 years, namely 2014-2018, was ranked 3rd after rice and corn so that it was easy to obtain. Edible films made from starch have the advantage of being able to protect products from oxygen, carbon dioxide, and lipids so that they can prevent dehydration and fat oxidation and make the product durable. However, starch-based edible films have several drawbacks, including being easily torn and hydrophobic, so that other additives such as plasticizers or plasticizers are needed.

A plasticizer is an additional material that can reduce the intermolecular forces in the polymer chain so that it increases elasticity and is not easily torn [3]. The type of plasticizer that is often used in the manufacture of edible films is glycerol. Glycerol (1,2,3-propanetriol) is a compound that has three hydroxyl groups (trivalent alcohol). In addition, glycerol is easily soluble in water, can bind water, and reduce the rate of water vapor [5]. Glycerol can be obtained from the transesterification reaction of used cooking oil, this is because glycerol can be found in the main composition of the oil in the form of glycerin which has been combined with stearic acid, oleic acid, palmitic acid, and lauric acid. Used cooking oil is household waste derived from coconut oil which has been used for frying many times and causes the acid content in it to be more saturated. If consumed, it can be harmful to health. Generally, waste cooking oil will be brownish-black in color due to the release of triglyceride bonds that form glycerol and fatty acids. Therefore, to reduce environmental pollution from waste cooking oil, it can be processed into a source of glycerol. Glycerol from the transesterification of waste cooking oil is a colorless liquid and has a slightly sweet taste [6].

According to Basuki [1], in their research on the characteristics of edible films from sweet potato starch and glycerol. The study used variations in concentrations of 5, 10, and 15% (w/w) glycerol and variations in sweet potato concentrations of 1, 2, and 3% (w/v). The results showed that the optimum edible film characteristics were obtained when the concentration of sweet potato was 3% (w/v) and glycerol 15% (w/w) with a yield of 55.567%, thickness 0.041 mm, tensile strength 26, 594 Mpa, elongation rate 56.59%. water vapor transmission 0.147 g.mm/m2.hour and moisture content of 11.947%.

Based on the description above, edible films made from sweet potato starch and glycerol can be used as environmentally friendly coffee packaging materials. Considering that coffee enthusiasts continue to increase and there are lots of instant coffee products on the market. Therefore, this study aims to make edible films based on sweet potato starch and glycerol from used cooking oil as a substitute for instant coffee plastic packaging.

RESEARCH METHOD

The method used in this research is a study of literature from various sources of national and international journals, this study compares the results of previous studies to know the basic ingredients of some starch and glycerol which have good physical and mechanical properties as edible films.

The ingredients used in the manufacture of this edible film include sweet potato starch, aquades, used cooking oil, NaOH, methanol, and commercial instant coffee. The manufacture of edible films is carried out in 2 stages, namely the manufacture of glycerol and the manufacture of edible films.

Making glycerol from cooking oil

First, the used cooking oil is allowed to stand for 1 day to settle the existing impurities and then filtered. Then added methoxide solution made from a mixture of 2 grams of NaOH and 100 ml of methanol. Mix the methoxide solution with used cooking oil in a ratio of 1:3 and heat at a temperature of 70-75°C while stirring at 80 rpm for 1 hour. The solution will separate into 2 layers, namely the top layer in the form of glycerol and the bottom layer in the form of biodiesel. The two layers are separated by decantation or by a separating funnel.

Edible Film Making

As much as 3 grams of sweet potato starch were weighed and dissolved in 100 ml of distilled water. Sweet potato starch solution was added with glycerol with various concentrations of 10, 20, and 30% (v/w). The solution was then stirred at 400 rpm and heated at 85°C for 10 minutes until it became a suspension. The suspension is then poured onto a glass plate measuring 15x7x1.5 cm. Then put in the oven for 15 hours at 45°C. After drying, the edible film is removed from the oven and cooled at room temperature for 30 minutes to make it easier to remove from the mold.

Observation

Observations made in the manufacture of this edible film were physical observations including moisture content, thickness, water vapor transmission rate (WVTR), and observations of mechanical properties including elongation and tensile strength.

RESULTS AND DISCUSSION

One of the materials that are often used in the manufacture of edible films is starch, which is a hydrocolloid group that is easy to obtain and economical. Starch is composed of amylose and amylopectin polymers. Oktavia [7] stated that the amylose content in sweet potatoes was 40% n-butanol with a yield of 46.69%, while amylopectin was 30% n-butanol with a yield of 52.25%. Determination of amylose content in sweet potatoes by UV-Vis spectrophotometry method showed 31.86%, while the amylopectin content was 58.95%. The role of amylose in the film to be made is that it will affect the texture of the film. The lower the amylose content, the harder and stiffer the film will be stated that the amylose content in sweet potatoes was 40% n-butanol with a yield of 46.69%, while amylopectin was 30% n-butanol with a yield of 52.25%. Determination of amylose content in sweet potatoes by UV-Vis spectrophotometry method showed 31.86%, while the amylopectin content was 58.95%. The role of amylose in the film to be made is that it will affect the texture of the film. The lower the amylose content, the harder and stiffer the film will be [8]. Then the amylopectin content also has expanding granular properties and good binding power, so it is often used as a raw material for gelatin replacement [7]. Further [3] stated that amylose content both have different functions, amylose is responsible for the formation of a thin film and the formation of a strong gel. Therefore, the levels of amylose and amylopectin in sweet potato starch are suitable for making edible films. However, the weakness in making edible films from starch is that the resulting film is not strong enough and easily brittle, so it is necessary to add other materials such as plasticizers.

The most commonly used plasticizer is glycerol. The content of glycerol can be obtained from waste cooking oil. Glycerol can be obtained from the transesterification reaction of used cooking oil, this is because glycerol can be found in the main composition of the oil in the form of glycerin which has been combined with stearic acid, oleic acid, palmitic acid, and lauric acid [6]. Based on research of [9] with the use of 70 mL of 2M NaOH and 100% excess ethanol, 303.35 mL of glycerol was obtained. The continuation of the writing from [9] that the glycerol will get bigger along with the larger the volume of NaOH and excess ethanol. The mechanism that occurs in the transesterification reaction of used cooking oil is shown in Figure 1.

$$R^{2} \longrightarrow R^{1} \longrightarrow R^{2} \longrightarrow R^{2$$

FIGURE 1. Transesterification Mechanism

The transesterification reaction is an equilibrium reaction, in which the alcohol group of the ester is replaced with another alcohol in a process similar to hydrolysis. This happens because the hydrolysis reaction does not use water but uses alcohol. Used cooking oil (triglycerides) when reacted with an alcohol, will produce an ester and glycerol. The transesterification reaction will run slowly. Therefore, a catalyst is usually added. The catalyst that is often used is NaOH or KOH. Catalysts are used to speed up slow reactions [10]. The cooking oil transesterification reaction is shown in Figure 2.

FIGURE 2. Cooking Oil Transesterification Reaction

The use of glycerol from used cooking oil is applied in the manufacture of edible films as has been done by [1] in his research on the characteristics of edible films from sweet potato starch and glycerol. The study used variations

in concentrations of 5, 10, and 15% (w/w) glycerol and variations in sweet potato concentrations of 1, 2, and 3% (w/v). The results showed that the optimum edible film characteristics were obtained when the concentration of sweet potato was 3% (w/v) and glycerol 15% (w/w) with a yield of 55.567%, thickness 0.041 mm, tensile strength 26, 594 Mpa, elongation rate 56.59%. water vapor transmission 0.147 g.mm/m2.hour and moisture content of 11.947%. In a study conducted by [11] it was explained that the addition of glycerol plasticizer can affect the physical and mechanical properties of the edible film produced, namely in the form of water content, thickness, tensile strength, elongation, and WVTR. The results of the analysis of the effect of glycerol concentration can be seen in Table 1.

TABLE 1. Average moisture content.	thickness, WVTF	L elongation and tensile strer	igth of sweet potato edible films

Concentration Glycerol	Water Content (%)	Thickness (mm)	WVTR (g/m2)	Elongation (%)	Tensile strength (N/mm2)
10%	12,50 ± 1,41 a	0.06 ± 0.0009 a	$1,79 \pm 0, 12 a$	8,75 ± 2,5 a	$0,75 \pm 0,17$ a
15%	$15,60 \pm 2,72$ ab	$0,06 \pm 0,0025$ a	$2,08 \pm 0,48$ a	$10,00 \pm 0,0 \text{ b}$	$0,28 \pm 0,01 \text{ b}$
20%	$17,61 \pm 2,05$ bc	$0,07 \pm 0,0080 \text{ b}$	$2,30 \pm 0,52$ a	$12,50 \pm 5,0$ bc	$0.13 \pm 0.01 \text{ b}$
25%	$20,80 \pm 2,63$ c	$0,09 \pm 0,0120$ c	$2,35 \pm 0,70$ a	$17,50 \pm 5,0 \text{ b}$	$0.07 \pm 0.03 \text{ b}$

Based on the results of the analysis of the study, it was shown that the addition of glycerol had a significant effect (P<0.05) on the water content of the edible film. The water content of the edible film will increase as the glycerol concentration increases. This is because glycerol can increase the coherence between molecules so that the amount of water bound to the hydrocolloid will increase. In addition, starch is a hydrophilic polymer so that the more the OH group of glycerol increases the amount of water-bound. Basuki [1] states that glycerol has the property of slightly evaporating so that when heated at the same time and temperature, the amount of glycerol available is only slightly reduced and can bind water in large amounts so that the water outside the granules becomes inside the granules and is no longer free thus increasing the water content edible film [11].

Huri and Nisa's [12] research results revealed that the addition of glycerol made the edible film thicker. Glycerol will bind to starch to form a starch-glycerol-starch polymer replacing the bond between starch and starch so that the thickness increases as the concentration of glycerol in the film paste increases. The addition of glycerol gave a significant effect (P<0.05) on the physical properties of the sweet potato edible film. The increase in the volume of the film solution that is poured into the glass mold is the same so that with the increase in the concentration of starch and glycerol, the total solids in the film after drying will increase and the starch pores will be closer together, increasing the density of the film layer [1].

In research conducted [11] showed that the addition of glycerol did not have a significant effect on the value of the water vapor transmission rate (WVTR). The results of the analysis showed that the highest steam transmission rate of sweet potato starch edible film ranged from 1.79 g/m2 to 3.33 g/m2. The highest water vapor transmission rate was produced at the addition of 25% glycerol while the lowest was at 10% glycerol concentration. The difference in the rate of water vapor transmission is not too significant due to the nature of glycerol which can increase the permeability of the film to air and water vapor if the concentration given is too high. The increase in water vapor transmission rate is due to the hydrophilic nature of glycerol and can reduce internal hydrogen bonds and increase the intermolecular distance which causes an increase in film mobility resulting in water transfer. Edible films that have a small water vapor transmission rate are suitable for packaging products that have high humidity. The edible film will inhibit the amount of water vapor released by the product to the outside so that the product does not dry quickly and on the contrary, an edible film can protect the water vapor that enters the environment. The value of the water vapor transmission rate has an inverse relationship with the thickness. Film thickness describes the distance traveled by water vapor to diffuse through the film. The thicker the edible film, the longer the distance traveled by the water vapor to diffuse [3].

In addition to affecting the rate of water vapor transmission, the addition of glycerol also affects the elongation of the edible film. According to [12] the addition of a plasticizer increases the elongation value of the edible film. The increase in elongation is because glycerol can interact with the starch polymer matrix by forming starch-

glycerol bonds. The hydroxyl groups along the glycerol chain will form hydrogen bonds between starch polymers replacing hydrogen bonds between starches. Glycerol can effectively reduce the internal hydrogen bonding thereby increasing the free space between molecules which causes a decrease in stiffness and an increase in film flexibility. The space will be filled with glycerol so that it will reduce the interaction tension between starch molecules.

Tensile strength is an important parameter for edible films as a substitute for packaging. Tensile strength is the maximum level of strain that can be maintained by the edible film before breaking. The addition of glycerol concentration in edible films resulted in a decrease in the water holding capacity between molecules because glycerol which has soluble properties will dissolve in polymer chains and facilitate the movement of polymer molecules to reduce the transition temperature on the product surface. If the transition temperature decreases, the tensile strength will increase. The tensile strength can be caused by attractive forces between the same particle (cohesion) and different particles (adhesion) namely glycerol. Glycerol is a plasticizer that has a low molecular weight and is easily soluble, so it easily enters the starch polymer matrix which can increase the flexibility of edible films [13]. The addition of glycerol that is too high can reduce the intermolecular tension that composes the film matrix so that the edible film will be weaker against mechanical treatment. The more polysaccharides in the edible film it will increase the stretching strength. This is because the amylose contained in the edible film will form hydrogen bonds, the higher the amount of amylose will increase the retrogradation properties of the suspension of the edible film after being heated so that it will increase its tensile strength [11]. Tensile strength according to JIS 1975 standard (Japanese Industrial Standard) is 0.39226 MPa.

Application of edible film as instant coffee packaging according to [3] The use of edible film from peanut husks shows that the use of edible film as coffee packaging can reduce the water content of instant coffee. The water content in instant coffee is a very important parameter because with increasing storage time, the water content will increase. The water content that continues to increase will cause instant coffee to spoil quickly which is characterized by product clumping. In addition, the water content can affect the Aw value and product stability during storage. Generally, roasted coffee or instant coffee has an Aw value ranging from 0.1-0.3 with a moisture content of not more than 4% at 20°C. The results of this study can be seen in Table 2.

No.	Sample	Water Content Without Edible Packaging 0 hours	Water Content Without Edible Packaging 24 hours	Water Rate With Edible Packaging 24 hours
1	c1p2	3,5%	8%	6%
2	c1p3	3,5%	8%	6%
3	c2p2	3,5%	8%	5%

TABLE 2. Results of Analysis of Water Content in Instant Coffee

These results indicate that the value of water content in instant coffee without using edible film tends to increase during storage time. The increase in water content is due to the permeability of the packaging material to water vapor. Increased permeability will make more water vapor from the environment that can pass through the packaging material. Instant coffee contains hygroscopic ingredients so it tends to absorb the steam from the air and is also influenced by the level of environmental humidity.

CONCLUSION

Based on several studies, it can be concluded that edible films can be made from sweet potato starch and glycerol obtained from waste cooking oil through a transesterification reaction. The addition of glycerol will affect the physical and mechanical properties of the edible film. The higher the concentration of glycerol added, the value of elongation, WVTR, thickness, the water content will increase. If the amount of glycerol added is within optimal limits, the resulting edible film will be stronger and not brittle. However, if the addition of glycerol is too high, it will reduce the properties of strong pull. The application of edible film as edible packaging for instant coffee can reduce the water content of instant coffee with a range of 1.5% lower than instant coffee that does not use edible packaging.

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