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Effect of the Safety of Chitosan Additive to Biodegradable Plastic Quality Based on Cellulose of Bacterial Glycerol from Coconut Water (*Cocos Nucifera*)

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Abstract:

Plastics are the most widely used polymer in life and cause global plastic pollution in the marine environment. Therefore research was conducted with the aim of making biodegradable plastics by utilizing bacterial cellulose produced by *Acetobacter xylinum* coconut water media with the addition of plasticizers namely 3.5% glycerol in the fermentation medium and soaking using additives namely chitosan with a concentration of 2%, 4%, 6%, and 10% on bacterial cellulose glycerol that has been formed. Bacterial cellulose fermentation was carried out to form a thickness of 0.5-1 cm and soaking bacterial cellulose glycerol in chitosan for 3 days. Plastic cellulose glycerol bacterial chitosan produced was characterized using water content test, tensile strength test, and crystallinity test (X-Ray). The test results of water content on chitosan bacterial cellulose glycerol plastic, the higher the concentration of chitosan, the smaller the percentage value of water content. The mechanical test results showed an increase in the value of tensile strength with the higher concentration of chitosan used. Analysis of the degree of crystallinity shows that immersion using chitosan can reduce crystalline percentage.

Keywords — Biodegradable Plastic, Coconut Water, Bacterial Cellulose Glycerol, Cellulose Glycerol, Bacterial Chitosan, Chitosan.

I. INTRODUCTION

Plastics are the most widely used polymer in our daily lives, especially in packaging applications [1]. In fact, 34 million tons of plastic waste is produced annually throughout the world and 93% of them are disposed of in landfills and oceans [2]. Plastic consumption in developing countries has been reported to be more than the world average due to higher levels of urbanization and economic

development. For example, developing countries such as China, Indonesia, the Philippines, Sri Lanka and Vietnam are reported to produce more than 50% of global plastic pollution in the marine environment [3].

New biodegradable films made from biopolymers play an important role in reducing the environmental impact of non-biodegradable plastic waste [4]. Biodegradable plastics or bioplastics are

plastics that can be used like conventional plastics, but will break down by the activity of microorganisms after they are used up and disposed of into the environment. Usually conventional plastics are made from petroleum, natural gas, or coal. While bioplastics are made of renewable material, namely from compounds contained in plants such as starch, cellulose, collagen, casein, proteins or lipids contained in [5].

Bacterial cellulose is one of the nanostructured biomaterials, featuring unique properties with a broad perspective for applications in various fields, including composite membranes, drugs, artificial skin, blood vessels, and binding agents [6]. There are several microorganisms that are capable of synthesizing cellulose, however *Acetobacter xylinum* is the only one that is quite a lot in industrial applications. The structure and mechanical properties of bacterial cellulose differ from the structure of plant cellulose, although the chemical composition is identical. Bacterial cellulose has high mechanical properties such as tensile strength and modulus, water retaining capacity, moldability, crystallinity, and biocompatibility [7].

As for making bacterial cellulose as biodegradable plastic can be done by utilizing coconut water waste. Where coconut water is rich in nutrients, namely sugar, protein, and fat, so it is very good for the growth of food product-producing bacteria. Young coconut water contains 95.50% water, 0.10% protein, fat less than 0.10%, carbohydrates 4.00%, and ash 0.40%. Young coconut water also contains vitamin C of 2.20 -3.40 mg / 100ml and vitamin B complex consisting of nicotinic acid, pantothenic acid, biotin, folic acid, vitamin B1, and a little pyridoxine. Young coconut water also contains a number of minerals, namely nitrogen, phosphorus, potassium, magnesium, chlorine, sulfur, and iron [8].

Coconut water is very easy to find and can be processed into another alternative, namely as a bacterial cellulose based biodegradable plastic which will produce quite a lot of benefits. To improve the properties of plastic it is necessary to

add plasticizers and additives. Plasticizers and additives to be used are glycerol and chitosan.

Glycerol is the simplest glyceride compound, with hydroxyl which is hydrophilic and hygroscopic. Glycerol is included in one of the alkyl trihydroxy compounds (Propra -1, 2, 3- triol) $\text{CH}_2\text{OHCHOHCH}_2\text{OH}$. It is found in almost all animal fats and vegetable oils as glycerin esters of palmitic, oleic, stearic and other fatty acids. Glycerol is a neutral compound, with a colorless sweetness, thick liquid with a melting point of 20°C and a high boiling point of 290°C , glycerol can dissolve completely in water and alcohol, but not in oil. Conversely, many substances can be more soluble in glycerol than in water or alcohol. Therefore glycerol is a good solvent.

Chitosan is a linear polymer of glycosidic β -1,4 related to 2-amino-2-deoxyl-D-glucopyranose, which makes it an excellent film-forming material. However, functional groups such as hydroxyl and amine chitosan are very good hydrogen bond donors and acceptors. There are intramolecular and intermolecular hydrogen bonds which are very strong in chitosan, making it a crystalline or semicrystalline material with various allomorphs. As a result, films made from pure chitosan are stiff and brittle. Chitosan has several beneficial properties including hydrophilicity, biocompatibility, degradability, anti-bacterial properties, and has a great affinity for enzymes. Chitosan is hydrophilic, holds water in its structure and forms a gel spontaneously, so chitosan easily forms a membrane or film. Biocompatible is defined as the ability of a material to give a biological response that is good (non toxic) and does not have carcinogenic properties [9].

The purpose of this study was to determine the effect of chitosan concentration variations on the manufacture of bacterial cellulose glycerol based biodegradable plastic from coconut water and determine the characteristics of physical properties, mechanical properties, and crystallinity of bacterial cellulose glycerol based biodegradable plastic from coconut water by immersing chitosan additives.

II. RESEARCH METHODS

A. Preparation of Bacterial Cellulose

Bacterial cellulose was produced from 600 ml coconut milk, 60 g sucrose, 6 g of urea and 21 mL glycerol were heated to boiling. Then this mixture in acidify with 25% acetic acid to a pH of 4-4.5. Still hot mixture was poured into a sterile container and covered with newspaper until it reaches a thickness of ± 1 cm.

B. Purification of Bacterial Cellulose

Bacterial cellulose washed with running water for ± 24 hours. And submerged in NaOH 2% for ± 24 hours. After soaking, cellulose washed again with running water and stored until BGC used.

C. Preparation of Chitosan Solution

Chitosan is weighed according to the variation of the required concentration of 2%, 4%, 6%, 8% and 10% then dissolved with 1% acetic acid as much as 600 ml.

D. Preparation of Cellulose Glycerol Bacterial Chitosan

Bacterial cellulose is cut to size and 15x2x1 2x2x1. Then immersed in 300 mL of Chitosan solution for 3 days in the shaker.

E. Immersion Cellulose Glycerol Bacterial Chitosan

cellulose which has been cut according to the required size and then soaked in chitosan solution 2%, 4%, 6%, 8%, and 10% and in the shaker for 3 days.

F. Testing the Characteristics of Cellulose Glycerol Bacterial Chitosan

The characteristics of cellulose glycerol bacterial chitosan obtained is tested with some parameters, namely :

1) Water Content

Test the water content of bacterial cellulose glycerol where cellulose was cut to size 4 cm x 2 cm, in weight with analytical balance. Then cellulose glycerol bactericidal chitosan is placed in the vaporizer and in the oven until dry, then weigh the plastic again. The Percent of Water contained can be calculated by using the equation:

$$\text{Water content \%} = \frac{a-b}{a} \times 100\%$$

Where:

a = initial weight (gram)

b = dry weigh (gram)

2) Tensile Strenght

The bacterial glycerol cellulose used has a size of 2x2x1 cm. then pressed using glass until it is thin and heated with a temperature of 105 to dry. after that, the sample was tested using a tensile strenght . from the results of the test can be obtained the value of tensile strength (stress) dan strain. While the elasticity value is obtained from the calculation, namely :

$$\text{Elasticity} = \frac{\text{stress (MPa)}}{\text{strain}}$$

3) Crystallinity of Plastics

Crystallinity analysis was carried out using X-ray Diffraction (XRD). Plastics cut in size 1 cm x 1 cm are then inserted into a sample template coated with wax. The degree of crystallinity is determined using the following equation:

$$\text{Crystalline degree\%} = \frac{a}{\text{wide area (a + b)}} \times 100\%$$

Where :

a = wide crystalline area (gram)

b = amorphous(gram)

II. RESULTS AND DISCUSSION

a) Testing of Water content

The water content test aims to determine the percentage of the amount of water contained in chitosan bacterial cellulose glycerol. Water content is an important parameter to determine the effect of plasticizing water on biopolymer films. The effect of cellulose glycerol bacterial chitosan soaking on the percentage of water content can be

seen in the following figure:

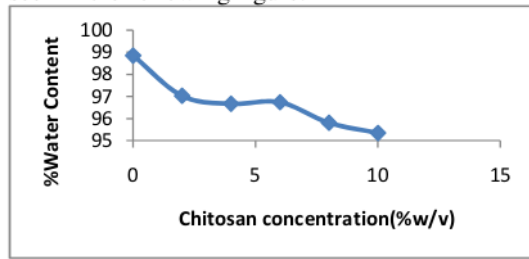


Figure 1. Effect of cellulose glycerol bacterial chitosan soaking on water content

In Figure 1, it can be seen that the water content decreases with the higher concentration of chitosan used to soak bacterial cellulose glycerol. The highest water content is at 0% SGBK which is equal to $\pm 98.85\%$, the percentage of water content decreases with soaking using chitosan where the lowest percentage of water content is at 10% SGBK of $\pm 95.35\%$. This happened because the water content contained in bacterial cellulose glycerol was replaced by chitosan, because the concentration of chitosan was higher than the concentration of water contained in bacterial cellulose glycerol. This is in accordance with the theory of osmosis, namely the transfer of solvents from higher concentrations to lower concentrations.

b) Tensile Strength

Tensile strength is the maximum pull that can be achieved until the film can survive before breaking. Tensile strength testing is done to determine the maximum force that can be held by a plastic. The effect of chitosan immersion on the tensile strength of SGBK plastic can be seen in the following figure.

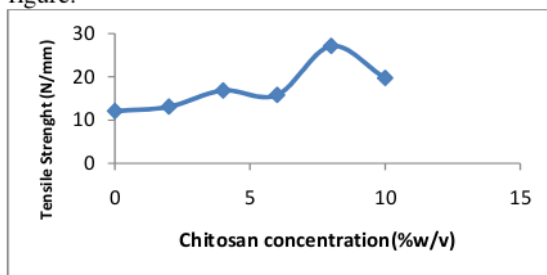


Figure 2. Effect of chitosan immersion on the plastic tensile strength of Glycerol Cellulose Chitosan Bacterial

Based on the image above, the plastic tensile strength of SGBK at the concentration of chitosan 0-4% increases and the highest tensile strength is obtained at a concentration of 8%. This is because hydrogen bonds are formed in plastic films. This hydrogen bond causes the plastic film to become stronger and harder to break [10]. But the concentration of 6% chitosan and 10% decreases the tensile strength. This is due to several factors such as unstable temperature and pressure when forming plastic sheets.

c) Degree of Crystallinity

The crystallinity test was conducted to determine the degree of crystallinity of chitosan bacterial cellulose glycerol plastic. This analysis was carried out using X-ray Diffractogram (XRD). X-ray diffraction can provide information about the structure of the polymer, including about the amorphous and crystalline state of the polymer. Polymers can contain crystalline regions that are randomly mixed with amorphous regions. The diffractogram of X-ray crystalline polymers produces sharp peaks, while amorphous polymers tend to produce widened peaks [11].

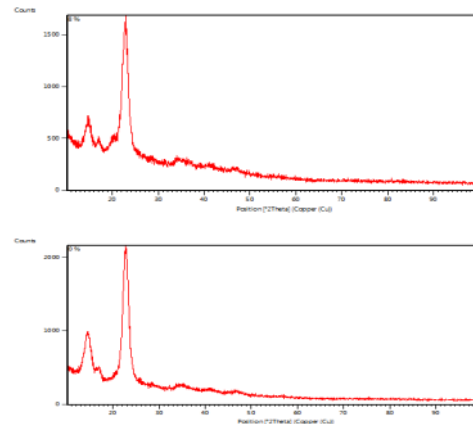


Figure 3 XRD Diffractogram: a) Bacterial Glycerol Cellulose and b) 8% Chitosan Cellulose-Glycerol.

Figure 3. shows a diffractogram of a) bacterial cellulose glycerol and b) 8% chitosan cellulose glycerol. Bacterial cellulose glycerol is a more crystalline material than chitosan bacterial cellulose glycerol. In the figure you can see the

dominant peaks appearing in the angle area 2θ between $20^\circ - 30^\circ$ from the XRD data. In 0% bacterial cellulose glycerol can be seen the peaks that appear in area 2θ , namely angles $14^\circ, 16^\circ, 22^\circ$, and 46° with peak intensity of 2000. In 8% chitosan bacterial cellulose glycerol plastic peaks that appear at area 2θ angle $14^\circ, 16^\circ, 20^\circ, 22^\circ$ and 34° with a peak intensity of 1500. Thus bacterial cellulose glycerol has a greater intensity than 8% chitosan cellulose glycerol.

Determination of cellulose crystallinity can be determined by several methods, namely Peakheightmethod, Ruland-Vonkmethode, Hermans-Weidingermethode, Jayme-Knollemethode, and Deconvolutionmethode. Based on the Hermans-Weidingermethode method used in determining the cellulose crystallinity degree, according to PH Hermans and A. Weidinger, the testing of crystallinity is done by copying the traces / photometer line traces in 2 copies on transparent paper or millimeter paper with known weight per unit surface, cut the picture, weigh the paper and take the average value. The percentage value of the degree of crystallinity of bacterial cellulose glycerol and 8% chitosan bacterial cellulose glycerol can be seen in table 1.

Table 1. % Degree of bacterial cellulose glycerol and 8% chitosan bacterial cellulose glycerol.

Sampel	m_{total} (gra)	m_{amorf} (gram)	$m_{kristal}$ (gram)	%Kristanilitas
SGB	0,0877	0,0126	0,0751	85,63
SGBK 8%	0,1045	0,0163	0,0882	84,40

Based on table 1. it was found that the percentage value of the degree of Christianity in SGB was 85.63% and the percentage of amorphous was 14.37% greater than SGBK which was 84.40% and amorphous percentage 15.60%. This proves that immersion using chitosan can reduce the crystallinity of plastic.

IV. CONCLUSION

The effect of immersion using chitosan as an additive to the manufacture of bacterial glycerol cellulose plastic can change the texture and color of bacterial glycerol cellulose, the higher the

concentration of chitosan used, the stiffer the chitosan bacterial cellulose glycerol produced. The effect of bacterial cellulose glycerol immersion using chitosan on water content is where it decreases with increasing concentration of chitosan used. Tensile strength has increased along with the higher concentration of chitosan used. The crystalline analysis showed that using chitosan immersed the percentage of chitosan bacterial cellulose glycerol to lower bacterial cellulose glycerol cellulose.

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