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The Effect of Additional Chitosan on the Manufacturing of Biodegradable Plastic from Coconut Water

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ABSTRACT: *Biodegradable plastic or bioplastic is a type of plastic that can be degraded naturally by microorganisms found in the environment. The purpose of this study was to determine the effect of adding chitosan to biodegradable plastics on physical properties, mechanical properties, biodegradation, and chemical structure. The use of chitosan variations used were 2%, 4%, 6%, 8%, and 10%. Based on the test results, it was found that the value of water content and swelling decreased with the addition of chitosan concentration. Optimum tensile strength and elasticity were obtained with the addition of 6% chitosan of 60.13 MPa and 825.61 MPa. The ability of plastic biodegradation decreases with increasing concentration of chitosan. The FTIR spectrum with the addition of chitosan and without the addition of chitosan showed the presence of O-H bonds at wave numbers 3500-3200 cm-1, C-H bonds at wave numbers 2800-2950 cm-1, C-O bonds at wave numbers 1000-1150 cm-1, and N-H bonds at wave numbers 1650-1580 cm-1. The degree of crystallinity decreased with the addition of chitosan to 65.24%.*

Keywords: *Coconut Water, Bacterial Cellulose, Biodegradable Plastic, Chitosan*

I. INTRODUCTION

Plastic is a polymer that is one of the most widely used in everyday life, for example, it is used as an ingredient in packaging [12]. In this day and age, the type of plastic that is most widely used by the public is a type of synthetic polymer plastic made from petroleum which cannot be decomposed by other microorganisms in the environment [4]. If things like this continue to happen, it can result in the accumulation of garbage which will cause pollution and damage to the environment [10]. Therefore, other alternatives are needed in the manufacture of plastics, namely plastics that are more environmentally friendly and the raw materials can be found from materials that are not difficult to obtain, namely biodegradable plastic.

Biodegradable plastic or bioplastic is a type of plastic that can be degraded naturally by microorganisms found in the environment [5]. Bioplastics are a family of polymer products with molecular structures that are susceptible to biological decomposition. In addition to the chemical structure of plastics, the level of degradation of biodegradable plastics is also controlled by environmental conditions such as temperature, humidity and nutrient content, all of which affect microbial activity [9].

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No	Characteristics	JIS Z1707	SNI Plastic
1.	Film thickness(mm)	0.25mm	-
2.	Tensile Strength (Mpa)	3,992 MPa	24.7-302
3.	Percent Elongation (%)	< 10% is very bad 50% very good	21-220
4.	Modulus of Elasticity	0.35 MPa	-

Figure 1. SNI for Bioplastics According to JIS Z1707 and SNI for Plastics

Synthetic materials are increasingly being used because of their relatively low cost, reproducibility, resistance to physical aging and biological attack. However, the use of synthetic polymer materials is only used for a short period of time and then disposed of into the environment which will cause accumulation, ideally the material should be biodegraded so that there is no accumulation of garbage in the environment [9].

Biodegradable plastic made from coconut water with Polyethylene Glycol (PEG) plasticizer has the disadvantage that it has weaker barrier properties against water, because PEG has hydrophilic properties [2]. The disadvantages of biodegradable plastic from coconut water with PEG plasticizer can be overcome by adding materials that have hydrophobic properties such as chitosan [2].

The addition of chitosan to biodegradable plastic is used because chitosan has the following properties: hydrophobic, biocompatible, can decompose, anti-bacterial and has a high affinity for enzymes. It is hydrophobic because chitosan has a structure that can hold water and can form gels easily, making it easy for membranes or films to form. Biocompatible means the ability of a material to provide a good biological response (non-toxic) and non-carcinogenic [7].

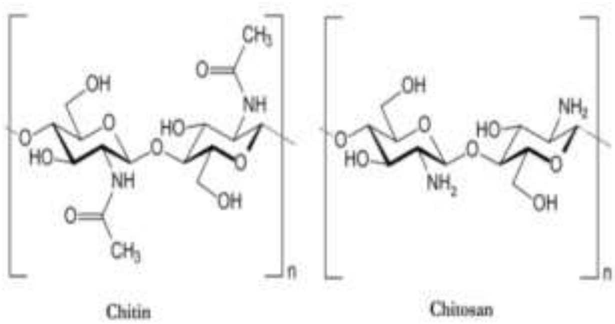

Figure 2. Chitosan Chemical Structure [21]

II. MATERIALS AND METHODS

2.1. Tools

The equipment in this study was divided into two, namely for sample preparation and characterization. The tools for sample preparation were glassware, plastic container measuring 24x17x4 cm, cooking pot, stove, rag, newspaper, tissue roll, filter, rubber, stirrer, knife, scissors, pH paper, analytical balance, iron, evaporating dish, and ovens. Equipment for characterization is Tenssion Testing (Universal Tensile Strength), FTIR, and XRD.

2.2. Material

This study used ingredients including aged coconut water from coconut milk sales waste, aquades, *A. Xylinum* inoculum, granulated sugar ($C_{12}H_{22}O_{11}$), food vinegar acid (CH_3COOH), urea fertilizer ($CO(NH_2)_2$), technical NaOH from bratachem, chitosan ($(C_6H_{11}NO_4)n$) from crustacean shell waste, and Polyethylene Glycol (PEG) 400.

2.3. Production of PEG Bacterial Cellulose

Put old coconut water in a saucepan and heat it until it almost boils, then add 600 g of sugar, 6 g of urea, 10 mL of PEG 400 14%, and 12 mL of acetic acid, then heat until it boils. After boiling, put the sample in a plastic container and let it sit until it reaches room temperature. After reaching room temperature, it was inoculated with A. xylinum starter, then fermented for 2 weeks until it reached a thickness of at least 0.5 cm.

2.4. PEG Bacterial Cellulose Purification

Bacterial cellulose that has reached its thickness should then be purified using 2% NaOH by soaking bacterial cellulose in 2% NaOH for 24 hours.

2.5. Synthesis of Biodegradable Plastics

Soak the bacterial cellulose using chitosan according to predetermined variations, namely 2%, 4%, 6%, 8%, and 10%. Then press the bacterial cellulose sheet using an iron between non-woven fabrics for 15 minutes which aims to reduce the water content in the plastic and prevent the plastic from breaking and shrinking.

2.6. Characterization of Biodegradable Plastics

a. Water Content Test (%WC)

Measurements were made by weighing the sample as the initial weight (W1), then in an oven at 105^{0}C until the weight was constant (W2).

$$\% \text{ WC} = \frac{\text{W1} - \text{W2}}{\text{W1}} \text{x } 100\%$$

b. Swelling Test

The results of the dry sample on the water content test were continued by immersing the plastic in 20 mL of water for 3 days, then weighing it until it weighed constant (W2).

$$\% \text{ Swelling} = \frac{\text{W2} - \text{W1}}{\text{W1}} \text{x } 100\%$$

c. Tensile Strength Test

The tensile test is the maximum stress that the plastic can withstand when it is pulled before the plastic breaks [8]. The plastic sample is clamped at both ends using a tensile strength tool, then the tool is operated until the sample breaks.

$$\text{Tensile Strength (MPa)} = \frac{\text{F}}{\text{Ao}}$$

d. Breaking Strength Test (Elongation)

The tensile strength or elongation test is carried out with the same steps as the tensile strength test.

$$\% \text{ Elongation} = \frac{\text{strain at break (mm)}}{\text{initial length (mm)}} \times 100\%$$

e. Elasticity Test

Testing the elasticity of biodegradable plastic can be seen from the tensile strength test and plastic elongation. The value of tensile strength is directly proportional to the value of elasticity.

$$\text{Elasticity (MPa)} = \frac{\sigma}{\varepsilon}$$

f. Biodegradation Test

The biodegradable plastic was cut to a size of 5 cm x 5 cm and then weighed using an analytical balance to obtain the initial weight (W1). The plastic is buried in the soil to a depth of 15 cm for 12 days. Next, the sample was taken from the soil, cleaned using a tissue and weighed to obtain the weight of the buried plastic (W2) [16].

$$\% \text{ Weight (W)} = \frac{W1 - W2}{W1} \times 100\%$$

g. FTIR (Fourier Transform Infra Red Spectrophotometry)

Characterization of the structure of biodegradable plastics using the FTIR instrument aims to determine the functional groups and types of bonds contained in biodegradable plastics. Biodegradable plastic samples were characterized at a wave number of 4000 - 600 cm^{-1}.

h. XRD (X-Ray Diffraction)

Samples were cut to a size of 2 cm x 2 cm. The sample pieces are placed on top of the sample holder and inserted into the XRD tool. The monitor will produce a fractogram that can be used to determine the degree of crystallinity of biodegradable plastic samples.

III. RESULT AND DISCUSSION

3.1. Biodegradable Plastic

PEG bacterial cellulose samples that have been soaked in chitosan for 3 days are then pressed using an iron between non-woven fabrics at maximum heat for 3 day (15 minute). This is done to produce plastic sheets that do not break and shrink. The results of drying using an iron can be seen in Figure 3.

Figure 3. Biodegradable Plastic

3.2. Water Content

The water content test aims to determine the percentage of the amount of water contained in the bacterial cellulose sample. Moisture content is an important parameter to determine the effect of plasticizers on water content in biopolymer films. The effect of immersion using chitosan on the percentage of water content of the sample can be seen in Figure 4.

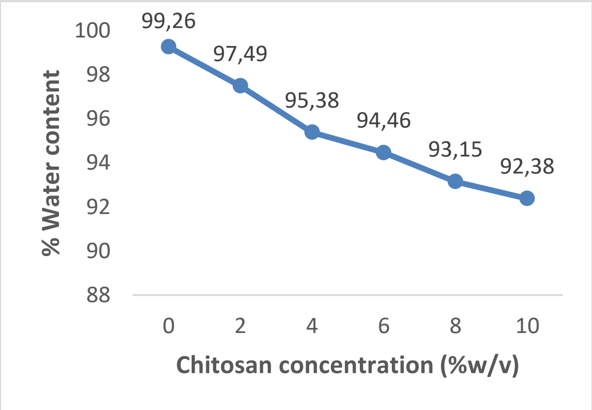

Figure 2. The Effect of Addition of Chitosan on the Water Content of Biodegradable Plastics

The water content will decrease with increasing concentration of chitosan used for soaking. The highest water content was found in bacterial cellulose without chitosan immersion which was 99.26% and the lowest water content was in bacterial cellulose with 10% chitosan immersion which was 92.38%. This can happen because the water content in bacterial cellulose without chitosan soaking is replaced by chitosan, because the concentration of chitosan is higher than the concentration of water contained in bacterial cellulose without chitosan immersion. This situation is in accordance with the theory of diffusion, namely the movement that occurs in the solvent from a higher concentration to a lower concentration [17].

3.3. Swelling

The nature of the resistance of bioplastics to water was determined by the swelling test, namely the percentage of swelling of the bioplastics in the presence of water. The longer the observation time, the higher the % swelling obtained due to the presence of water that diffuses into the bioplastic [14]. The results of the percentage of swelling due to immersion of biodegradable plastic in water can be seen in Figure 5.

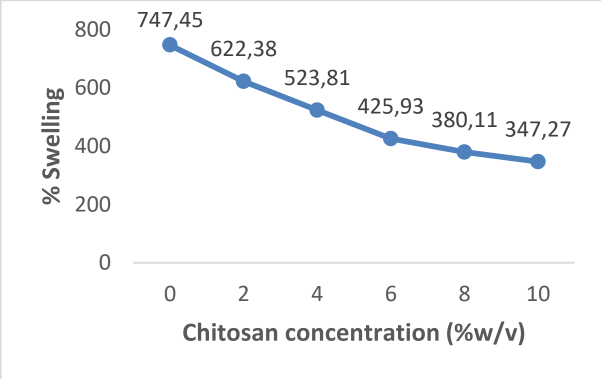

Figure 5. Effect of the addition of chitosan on the swelling of biodegradable plastics

The value of the degree of plastic swelling decreased with the addition of the concentration of chitosan. The result of the highest degree of swelling was in bacterial cellulose without chitosan immersion, which was 747.45% and decreased in bacterial cellulose with 10% chitosan immersion, which was 347.27%. Based on the results of these data, it can be concluded that the amount of water that can be absorbed by plastic soaked in chitosan will be less than plastic without soaking in chitosan. This can happen because chitosan is hydrophobic and insoluble in water. The water-absorbing properties of plastic films can also be seen in the morphological structure of plastic films which have cavities that function to absorb water content [6].

3.4. Tensile Strength

Tensile strength testing is measuring the tensile strength of plastic by applying a load until the plastic breaks or tears [18]. The tensile strength test is carried out to determine the maximum force that a plastic can withstand. The tensile strength test was carried out to determine the maximum force the plastic. The effect of immersion using chitosan on the tensile strength of biodegradable plastic can be seen in Figure 6.

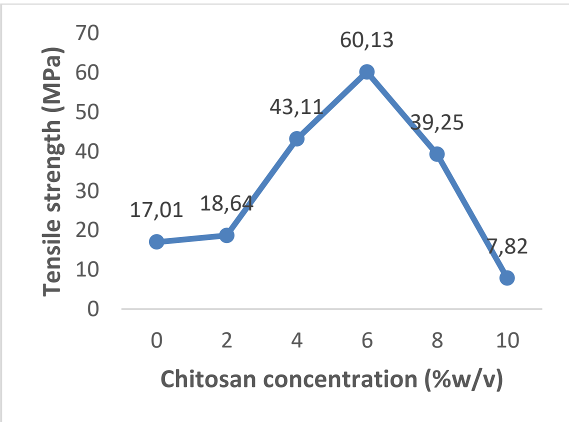

Figure 6. Effect of Addition of Chitosan on Tensile Strength of Biodegradable Plastics

The tensile strength value of bacterial cellulose without chitosan immersion to bacterial cellulose with 6% chitosan immersion is increasing and the highest tensile strength value is found in bacterial cellulose with 6% chitosan immersion, which is 60.13 MPa. This can happen because the greater the concentration of chitosan, the more hydrogen bonds contained in bioplastics so that the chemical bonds of bioplastics will be increasingly difficult to be strong and difficult to break, because it requires a large amount of energy to break bonds [14]. However, in 8% and 10% chitosan immersion bacterial cellulose, the tensile strength value decreased, due to the matrix in the plastic having passed its saturation point.

3.5. Elongation

Elongation is the maximum length value experienced by plastic when it is stretched or stretched until just before the plastic breaks [15]. The effect of soaking using chitosan on the elongation of biodegradable plastic can be seen in Figure 7.

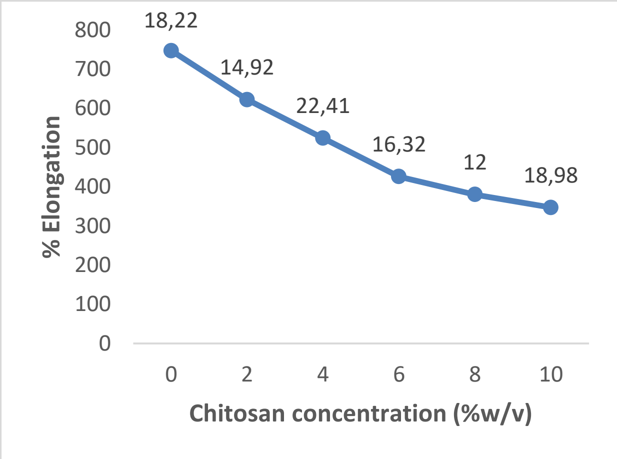

Figure 7. Effect of Addition of Chitosan on Elongation of Plastics

The lower elongation value with increasing concentration of chitosan used at the time of immersion. This happens because of the strong interaction between the mixture of cellulose and chitosan molecules. The molecular bonds of cellulose and chitosan that occur will be tighter if there is more chitosan so that the plastic will be difficult to stretch or elongate, this will certainly reduce the percentage level of elongation [13]. The lowest % elongation value was found in bacterial cellulose with 8% chitosan immersion by 12%, but in bacterial cellulose with 4% chitosan immersion there was an increase in the % elongation value, this was possible due to the unstable temperature factor during the process of forming the plastic sheet.

3.6. Elasticity

Elasticity is a measure of the stiffness of a material. If the elasticity value is higher, it means the material is getting stiffer [20]. The effect of chitosan immersion on the elasticity of biodegradable plastic can be seen in Figure 8.

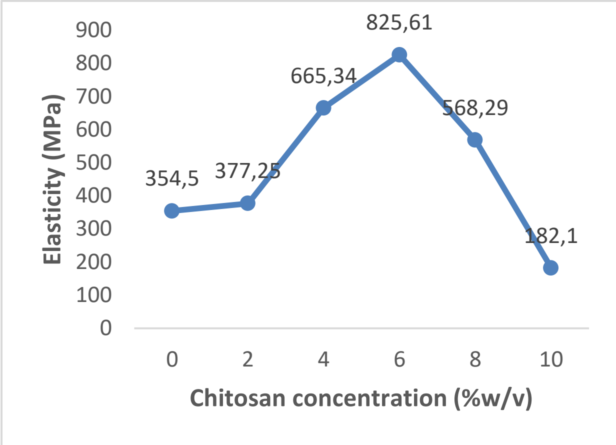

Figure 8. Effect of Addition of Chitosan on the Elasticity of Biodegradable Plastics

The elasticity value was higher along with the greater the concentration of chitosan used at the time of immersion to get the maximum value, namely in bacterial cellulose with 6% chitosan immersion of 825.61 MPa, then in bacterial cellulose with 8% and 10% chitosan immersion there was a decrease in elasticity value. , this also indicates that the elasticity has reached the optimum condition.

3.7. Biodegradation of Biodegradable Plastic

Biodegradation is the ability of microorganisms to break down or degrade natural polymers and synthetic polymers. Biodegradation testing is a test carried out to determine the level of resistance of biodegradable plastics to microbial decomposers, temperature, soil moisture, and physical and chemical factors in the soil. The effect of adding chitosan to the biodegradable plastic biodegradation process can be seen in Figure 9.

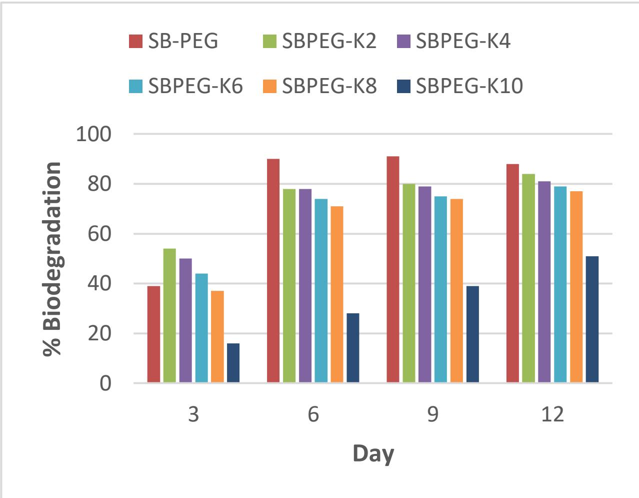

Figure 9. Effect of Addition of Chitosan on Biodegradation of Biodegradable Plastics

Plastics that are not soaked in chitosan and buried in the ground for 12 days will be more easily degraded than plastics soaked in chitosan. This is due to chitosan as a natural reinforcement which has hydrophobic properties, which is difficult to dissolve in water contained in the soil. Another cause is that chitosan has properties that are resistant to attack by decomposing microorganisms contained in the soil [6].

3.8. FTIR

The characterization of the functional groups contained in edibles using the FTIR instrument was carried out at a wave number of 4000 - 600 cm^{-1}.

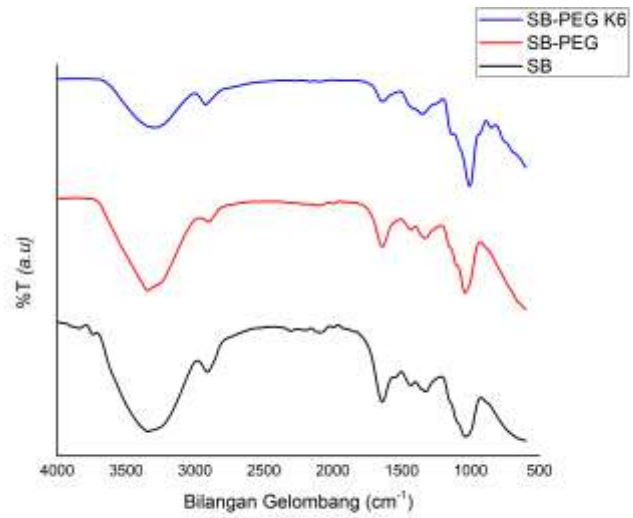

Figure 10. FTIR Spectrum of Biodegradable Plastics

The first spectrum (SB), the second spectrum (SBPEG) and the third spectrum (SBPEG-K6) showed the presence of O-H bonds at wave numbers around 3500-3200 cm^{-1}, C-H bonds at wave numbers around 3000-2840 cm^{-1}, C-O bonds at the wave number is around 1085-1050 cm^{-1}, and the NH bond which is the intensity of the N-H bond medium at the wave number between 1650-1580 cm^{-1}. Based on the functional group analysis test using FTIR, it shows that there is no new functional group formed. This shows that the process of making bioplastics accompanied by the addition of additives is a physical blending process [11].

3.9. XRD

The information obtained in the XRD analysis is in the form of a diffractogram graph that shows the peaks of the crystal structure of a material. The crystalline structure will produce sharp peaks, while the amorphous structure will produce broad peaks [19]. The diffractogram graph of the three bioplastic samples is shown in Figure 11.

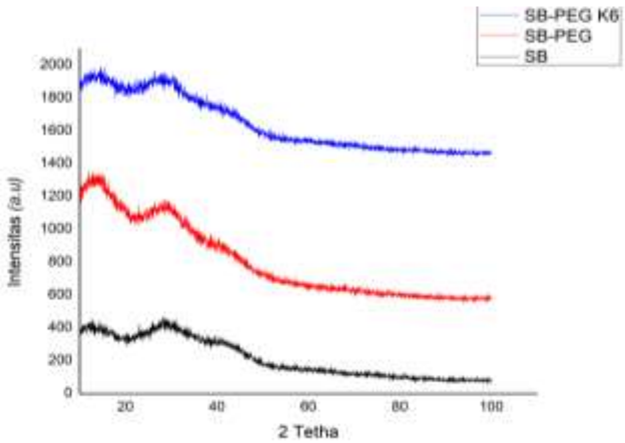

Figure 11. Biodegradable Plastic XRD Diffractogram

There are peaks of pure bacterial cellulose appear in the area 2θangles of 30° and 21°. According to Islami (2015) that the typical peaks of bacterial cellulose are located at 2θ, namely at 14°, 16°, 23°, and 34° with a percentage of crystallinity of bacterial cellulose is 75.47%. In PEG bacterial cellulose plastic the peaks that appear are at an angle between 14°-30° with a percentage of crystallinity is 93.25%, while in PEG bacterial cellulose plastic with 6% chitosan immersion the peaks that appear are at2θ angle between 20°-29° with a percentage of crystallinity is 65.24%.

IV. CONCLUSIONS

Based on the research that has been done, it is obtained conclusion:

1. The addition of chitosan additives to PEG bacterial cellulose can reduce the percentage of water content of PEG bacterial cellulose from 99.26% to 92.38%.
2. The addition of chitosan additives to PEG bacterial cellulose can reduce the swelling percentage of PEG bacterial cellulose from 747.45% to 347.27%.
3. Variations of chitosan additives can increase the tensile strength and elasticity of PEG bacterial cellulose at a concentration of 6%.
4. The addition of chitosan additives did not add new groups of PEG bacterial cellulose and the crystallinity properties of PEG bacterial cellulose decreased with the addition of chitosan.

V. ACKNOWLEDGMENTS

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