Effect of Carrageenan Addition on the Quality of Bacterial Cellulose-Based Biodegradable Plastic from Coconut Water

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Abstract

Plastic is often used in daily life, resulting in a lot of plastic waste. Therefore, a solution is needed, namely the production of biodegradable plastic. This research aims to see the effect of carrageenan addition variation on the quality of bacterial cellulose-sorbitol-based biodegradable plastic from coconut water (Cocos nucifera) which includes mechanical properties testing (tensile strength, elongation, and moduulus young), biodegradation testing, and testing with FTIR and XRD instruments. In this study, sorbitol 30 % was used as a plasticizer with carrageenan variations of 0%, 0.5%, 1%, 1.5%, and 2%. The mechanical properties test results obtained a maximum tensile strength value of 126.58 Mpa in the 2% carrageenan addition variation, where the tensile strength value has met SNI standards for synthetic plastics which ranges from 24.7-302 Mpa, the Modulus young value obtained is 980.46 Mpa, and the elongation value is 21.68%. For the biodegradation test, the plastic added with carrageenan increased and on the 15th day the plastic was completely degraded. On FTIR functional group analysis, no new groups were formed and on XRD analysis on the addition of carrageenan, the % crystallinity of the plastic was greater at 98.66% compared to bacterial cellulose 97.01%, and bacterial cellulose-sorbitol 95.79%.

Keywords: *Coconut Water; Carrageenan; Sorbitol; Biodegradable Plastic; Bacterial Cellulose*

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INTRODUCTION

Plastic is one of the objects that is light, cheap and durable so that almost every industrial sector uses plastic as packaging or as its basic material. Around 100 million tons of plastic are produced worldwide each year for use in various industrial sectors, which means the amount of plastic waste produced is also the same every year (Arini et al., 2017). This plastic waste causes problems for the environment because the plastic degradation period is getting longer and breaks down into microplastics (Nikolopoulou et al., 2023).Therefore, one of the ways needed to overcome the pollution that arises is to replace conventional plastic with plastic that is more easily broken down and comes from natural sources or is called biodegradable plastic. Biodegradable plastic is an environmentally friendly plastic that can break down in the soil as a whole so that it does not cause environmental problems.

There are several ways to biodegrade plastics. For example, blending natural components like cellulose, starch, or chitin might improve the biodegradability of polymers used in plastic products (Al-Salem et al., 2019). Polysaccharide-based materials are among those that can serve as a foundation for the development of biodegradable plastic. Cellulose is one of the polysaccharides utilized. One of the celluloses is bacterial cellulose. Bacterial cellulose is cellulose produced by acetic acid bacteria and has several advantages compared to plant cellulose. Its advantages are that it has high purity, excellent network structure and high degradation ability.

Acetobacter xylinum bacteria are bacteria that are commonly used in the production of bacterial cellulose because they can use other types of sugar, glucose or glycerol, to produce pure cellulose. The use of mature coconut water as the main medium for the fermentation process can be considered because it contains various types of nutrients needed for bacterial growth. Apart from that, old coconut water also contains high levels of glucose and can produce maximum biocellulose production (Sulaiman et al., 2018).

Plasticizers are very necessary in making biodegradable plastics. The plasticizer used in this research is sorbitol. Sorbitol can be used as a plasticizer because it can increase the flexibility of films or plastics by reducing hydrogen bonds between molecules and increasing the molecular distance between polymers along the polymer chain (Lim et al., 2020).

Table 1. SNI Plastic

No.	Characteristics	Plastic SNI
1	Film (Mm)	-
2	Tensile Strength (Mpa)	24,7-302
3	Elongas*i* (%)	21-220
4	Hidrofobisitas (%)	99
5	Modulus Young	-

Additives are also needed as additional materials to improve the quality of biodegradable plastic. The additive added is carrageenan. Carrageenan is widely used in the food industry and other industries because of its thickening, swelling, and stabilizing properties. Carrageenan can also improve the mechanical properties of bioplastics, resulting in higher tensile strength. (Khotimah et al., 2022). Carrageenan is widely used as an ingredient for making edible films. Based on research (Taftazani Anaka & Dewata, 2022) the results of carrageenan variations show that carrageenan can be used to increase the quality of Edible Film, but several tests have not met the test standards.

Based on the description above to increase utilization coconut water into biodegradable plastic and trying variations of carrageenan on biodegradable plastic based on bacterial cellulose for this the author wants to do research on "Effect of Carrageenan on the Quality of Biodegradable Plastic Based on Bacterial Cellulose - Sorbitol from Coconut Water *(Cocos Nucifera).*

RESEARCH METHODS

The research will be conducted from March to July 2024 at the Chemistry Department Laboratory, FMIPA, Padang State University. Equipment for the manufacture and characterization of bacterial cellulose are laboratory glassware, plastic container size 24x17x4 cm, cooking pot, stove, sieve, newspaper, rubber, stirrer, knife, scissors, ph paper, analytical balance, aluminum foil, oven, autoclave, erlemeyer, tenssion testing astm vol 03.10 in 1991, FTIR and XRD. The materials used in this study include old coconut water (coconut milk and coconut seller waste in Patenggangan), inoculum *A. xylinum* (nata de coco lima bersaudara siteba), sucrose, technical acetic acid, urea fertilizer, water, sorbitol 30%, NaOH 2%, distilled water and carrageenan.

Work Procedure

1. Sterilization of Carrageenan

Carrageenan variations were used, namely 0.5 g; 1 g; 1.5 g and 2 g which were dissolved with 100 ml of distilled water using a magnetic stirrer at a temperature of 70-80^{O}C. After that, the carrageenan solution was sterilized using an autoclave for 15 minutes which will be used to make carrageenan bacterial cellulose media.

2. Preparation of Bacterial Cellulose

600 mL of old coconut water, 60 grams of sugar, 6 grams of urea and heated to boiling, then added plasticizer sorbitol 30% as much as 10 mL, then add acetic acid to the pH range of 4.5-5.4 into the pot and heated to boiling. After boiling, it was transferred to a plastic container and then the additional sterilized carrageenan variations (0%; 0.5%; 1%; 1.5%; 2%) were covered using newspaper that had been sterilized in the with oven. Then the media was left until room temperature. After reaching room temperature, inoculated with starter *A. xylinum* in the ratio of 10:1 (%v/v). During inoculation, the container should not be shaken and fermented for 14 days at room temperature until bacterial cellulose of at least 0.5 cm is formed.

3. Washing and Purification of Bacterial Cellulose (SBS-K)

Bacterial cellulose that has been formed is washed with running water, then soaked with 2% NaOH (%w/v) for 24 hours. After that, it was washed again with running water until the NaOH is gone, characterized by a pH that is not base.

4. Preparation of SBS-K Plastic Sheet

The purified sorbitol-carrageenan bacterial cellulose. The purified bacterial cellulose sorbitol-carrageenan was cut according to the required size and then oven at 105^{o}C for 60 minutes. The plastic sheet is ready to be tested and characterized.

5. testing the mechanical properties of biodegradable plastics

a. Tensile Strength Test

Tensile strength was calculated with Tensile Strength Industries model SSB 0500. Analysis of plastic tensile strength is done through data obtained from the tensometer. The amount of tensile strength can be calculated by using the equation:

$$\sigma t = \frac{Fmaks}{Ao}$$

Where:
F Max = Force Exerted by the Tool (N)
Ao = Cross-Sectional Area (Mm^2)

σt = Tensile Strength (Mpa)

b. Elongation Test

The measurement of breaking strength also uses the same way as the Tensile strength test. Elongation is expressed as a percentage, calculated using the following equation:

$$\%\ \textbf{Elongasi} = \frac{\textbf{Stretch at Break(Mm)}}{\textbf{Initial Length}} \times \mathbf{100\%}$$

c. Modulus Young Test

The elasticity of biodegradable plastics is seen from the tensile strength test. tensile strength and sample stretch. Elasticity can be calculated by means of:

$$\boldsymbol{E = \frac{\sigma}{\varepsilon}}$$

Where:
E = Modulus Young (Mpa)
σ= Tensile Strength
Ɛ = Percent Elongation

d. Biodegradation testing of plastic sheets

Was done by burying the plastic sheet in the soil with a size of 2x 2 cm at a depth of depth of 5 cm. The measurement process was carried out for 15 days. Before burying, the plastic was weighed in mass, then buried in the ground for 15 days with a weighing interval of every 3 days. Weighing interval every 3 days. The decomposed plastic can be calculated using the following equation:

$$\%\ \boldsymbol{biodegradasi} = \frac{\boldsymbol{mo - m}}{\boldsymbol{mo}} \times \mathbf{100\%}$$

Where:
Mo = Mass of Sample Before Burial
M = Mass of Sample After Buried

6. Molecular Structure Analysis of Biodegradable Plastic

a. Functional Groups analysis with FTIR

FTIR will analyze the functional groups of SBS recorded using a spectrophotometer at room temperature. Before the test is carried out, the sample holder is cleaned first using 70% ethanol. The SB plastic sample is placed on the sample holder, and the device is ready for use. The fource gauge on the swing arm was set with a wave number of 90-100 cm-1 (for solid samples). The sample was moved with a wave number of 4000-400 cm-1. The resulting spectrum on the monitor will be analyzed to determine the functional groups in

SB plastic. The FTIR measurement results show a spectrum with a horizontal curve showing the number of waves and a vertical line showing the percent transmission.

b. Crystallinity Analysis of Plastics Using XRD

The plastic was cut to a size of 5 x 5 cm, then inserted into a sample template coated with wax. The degree of crystallinity (DK) was determined using the following equation:

$$\% \ \mathbf{DK} = \frac{\mathbf{height}}{(\mathbf{height} + \mathbf{low}\,)} \times \mathbf{100}\%$$

RESULT AND DISCUSSION

The purpose of tensile force testing is to determine the mechanical properties of bioplastics that can withstand tensile forces from heavy loads. Bioplastics which have high tensile strength can protect the products they package from mechanical (Danni et al., 2023). One of the most crucial tests in the production of plastic is the tensile strength test, which establishes the material's quality and verifies that it satisfies SNI standards.

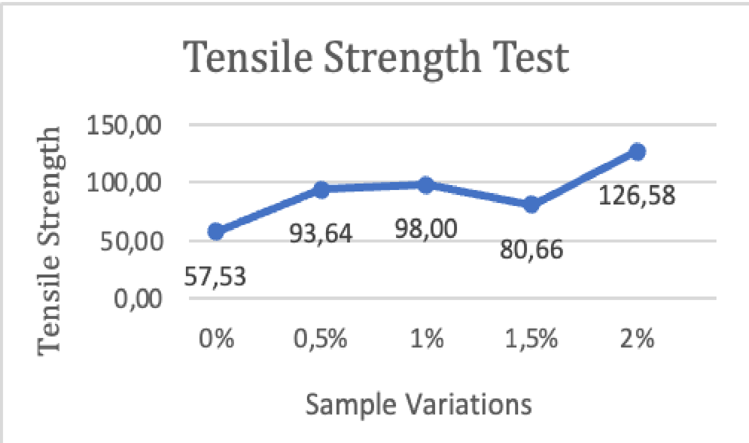

Figure 1. SBS-K Plastic Tensile Strength Graph

The graph above illustrates how the tensile strength value of SBS-K plastic increases at a carrageenan concentration of 1%, decreases to a concentration of 1.5%, and then increases again to a concentration of 2%. This occurs because the mixing procedure is less homogeneous, resulting in an uneven distribution of the plastic constituent parts. SBS plastic with the 2% carrageenan variant has the highest tensile strength value of 126.58 Mpa, while SBS plastic without carrageenan has the lowest tensile strength value of 57.53 Mpa. This is in line with research conducted by (Khotimah et al., 2022) stating that carrageenan can improve the mechanical properties of bioplastics.This is because the plastic has more hydrogen bonds when carrageenan is used at higher concentrations. As a result, chemical bonds become stronger and require more energy to break them (Marsa et al., 2023). In addition, the tensile strength values obtained were proven to meet the SNI criteria for plastic tensile strength. The best quality plastic is SBS-K 2% plastic which will be used for FTIR, XRD and biodegradation testing. This is determined by a tensile strength test.

A. Elongation Test

The elongation of biodegradable plastic is the percentage change in length that occurs when it is pulled until it breaks. To carry out this test, the increase in length that occurs before the tensile strength test is compared with that before carrying out the tensile strength test (Wahyunita et al., 2022).

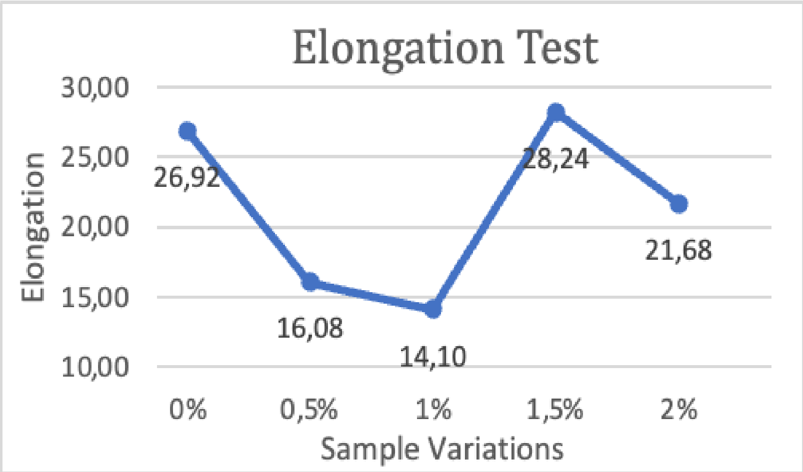

Figure 2. SBS-K Plastic Elongation Test Graph

Based on the graph, the plastic with the highest elongation value percentage was SBS-K plastic 1.5% at 28.24% and the plastic with the lowest elongation value was SBS-K plastic 1% at 14.10%. The higher the mass of carrageenan added will produce a film matrix that is stronger and less likely to break, thereby reducing the elongation value so it can be concluded that the elongation value is inversely proportional to the tensile strength value. The fact that the mixing was less uniform and the components in the plastic were distributed unevenly is one of the reasons why the elongation values for SBS-K plastic 1.5% increased rather than decreased.

B. Modulus Young Test

Modulus young is the ability of a material to return to its initial shape when the force applied to the object is stopped. The purpose of this test is to measure the stiffness of the biodegradable plastic produced.

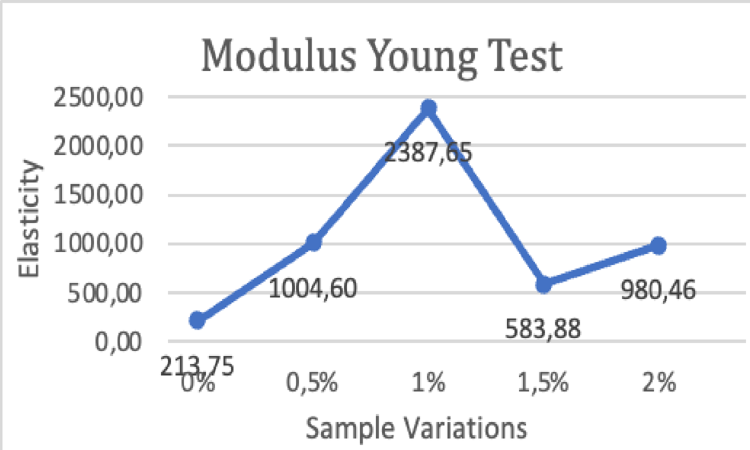

Figure 3. SBS-K Plastic Elasticity Test Graph

Figure 5 shows that the addition of carrageenan causes an increase in the modulus young value. However, the addition of carrageenan at a concentration of 1.5% caused a decrease in yield as shown in the modulus young graph. This is due to the less homogeneous mixing process causing the bacterial cellulose component to be distributed unevenly.

Modulus young is directly proportional to tensile strength, so that the greater the concentration of carrageenan used in the process of making biodegradable plastic, the thicker the number of polymers that make up the matrix, the greater the force required, resulting in a stronger tensile strength. Bioplastics with high tensile strength will be able to protect the products they package from mechanical disturbances well (Flury & Narayan, 2021).

C. Biodegradation Test

Degradation is the ability of microbes to convert compounds into carbon dioxide, water, methane, and inorganic components or biomass through enzymatic mechanisms that can be tested over a certain period. In this test, soil conditions were controlled at pH 7 and at room temperature.

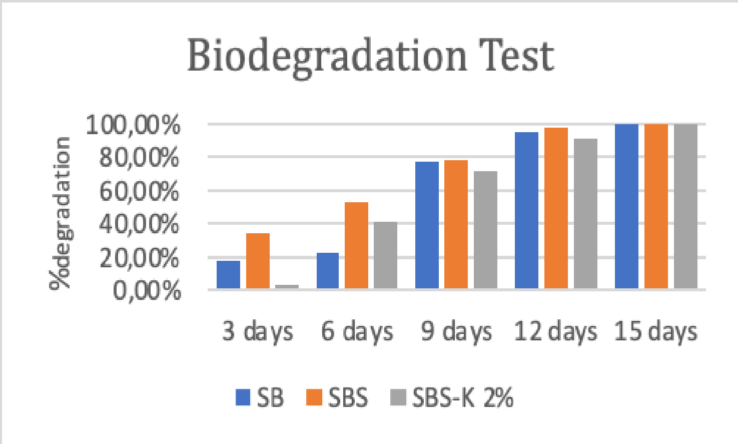

Figure 4. SBS-K plastic Biodegradation Test Graph

Figure 4 illustrates how the rate of plastic biodegradation can be slowed by increasing the concentration of carrageenan. However, on day 15, all the plastic was completely degraded.

Table 2. % degradation of SBS-K plastic

Day	SB	SBS	SBS-K 2%
3 days	17,54%	34,99%	3,09%
6 days	22,97%	53,50%	41,82%
9 days	77,91%	78,15%	71,41%
12 days	94,85%	97,52%	91,04%
15 days	100%	100%	100%

Based on the table it can be seen that the longest biodegradation in SBS-K 2% and the fastest in SBS without the addition of carrageenan. This is because the addition of carrageenan causes an increase in the thickness of biodegradable plastic which results in low water absorption in the plastic (Alfian et al., 2020). The higher the ability of plastic to absorb water, the faster its biodegradation will be. Biodegradation is also related to tensile strength, where the higher the tensile strength, the longer the biodegradation will take because of the hydrogen bonds that strengthen the plastic.

D. Characterization of Functional Groups of SBS-K Plastic

Functional group analysis using FTIR aims to determine the processes that occur during physical or chemical mixing, so that each sample in the process of making bacterial cellulose (SB) is analyzed using FTIR.

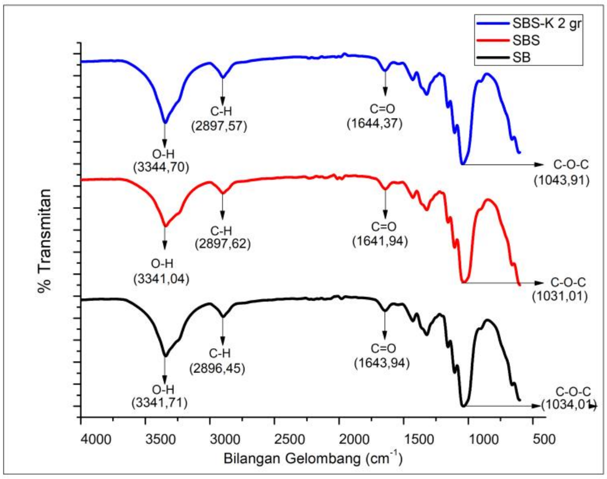

Figure 5. SBS-K Plastic FTIR Spectrum

Based on Figure 5, the functional groups found in the SB, SBS, and SBS-K samples are the O-H group at wave numbers between 3500-3200 cm^{-1}, the C-H bond is at wave numbers between 2850-2970 cm^{-1}, the C=O 1500-1675 cm^{-1}, C-O-C bond 1500- 1000 cm^{-1}. This is in accordance with the research (Rahmayetty et al., 2023) which states that bacterial cellulose has 4 main functional groups, namely O-H, C-O-C, C-H, and C=O. The results of functional group analysis testing with the FTIR instrument showed that only a shift in functional groups occurred and no new functional groups were formed. The addition of Sorbitol and carrageenan to SB causes a shift in this functional group. In the process of making biodegradable plastic accompanied by additives, a physical combination process is carried out so that no new functional groups are found (Hayati et al., 2020)

There are O-H and C-O-C eter functional groups which are hydrophilic, so they are able to bind air molecules in the environment, making plastic easily degraded more quickly by microorganisms. However, an increase in the number of SBS-K waves indicates an increase in vibrational energy, thereby increasing the tensile strength value and slowing down time degradation (Astuti et al., 2019).

E. Characterization of the Degree of Crystallinity of SBS-K Plastic

Crystallinity testing was carried out using a tool (XRD) with the aim of determining the degree of crystallinity of SB, SBS and SBS-K plastic samples.

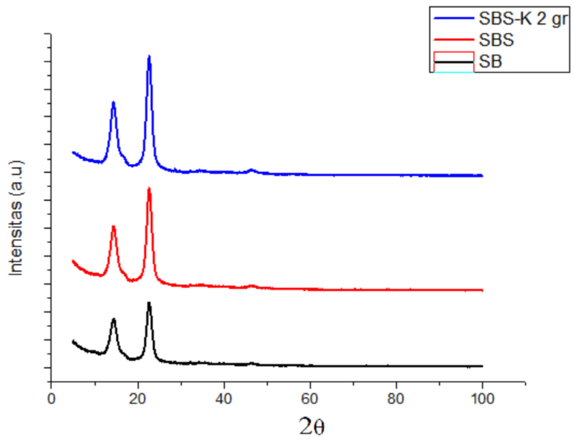

Figure 6. SBS-K Plastic XRD Diffractogram

Based on Figure 6, it can be seen from the diffractogram of biodegradable plastic that there are sharp and wide peaks indicating the presence of crystalline and amorphous.

Table 3. SBS-K Plastic Deggre of Crystallinity

Sample	**highest intensity**	**peak 2θ**	**height [cts]**	**low [cts]**	**% crystallinity**
SB	100	22,84	3624,15	108,25	97,01%
SBS	100	14,56	3159,21	133,06	95,79%
SBS-K 2%	100	22,73	6908,4	92,36	98,66%

Based on the calculation of the degree of crystallinity in each biodegradable plastic sample, it was found that the degree of crystallinity of pure SB plastic was 97.01%, so that pure SB plastic had an amorphous structure of 2.99%. The degree of crystallinity of SBS is 95.79% and the amorphous structure is 4.21% and for the degree of crystallinity SBS-K 2% has a degree of crystallinity of 98.66% and the amorphous structure is 1.34%.

The degree of crystallinity will affect the tensile strength and ability of biodegradable plastic to degrade. In the plastic biodegradation process, the higher the level of plastic crystallinity, the more difficult it is for the plastic to decompose. However, the amorphous part of biodegradable plastic will decompose more quickly because it is more susceptible to attack by decomposing microorganisms in the soil. ((Maneking et al., 2020). The part of the polymer that is less organized is amorphous because the amorphous part has higher functional groups that serve as the substrate of the crystalline. In addition, the degree of crystallinity in plastics determines the tensile strength value, which increases as the degree of crystallinity increases (Suryadinata & Putra, 2023).

CONCLUSION

Based on the results of the research that has been carried out, it can be concluded that the addition of carrageenan additives with various variations to biodegradable plastics has an influence on the tensile strength, elongation and elasticity values. The research results show that the addition of 2% carrageenan has the best quality of biodegradable plastic with a tensile strength of 126.58 Mpa, elongation of 21.68% and modulus young of 980.46 Mpa, where these results have reached SNI plastic standards. In the biodegradation test, SBS-K 2% took longer to degrade than SB and SBS, but all three were degraded 100% on day 15. In FTIR characterization no new functional groups were formed. The degree of crystallinity of biodegradable plastic increased after adding carrageenan.

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