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Bacterial Cellulose-Based Biodegradable Plasticfrom Pineapple (Ananassativus) Skin Waste: The Effectof Sorbitol On The Quality Of The Biodegradable plastic

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Abstract. The aim of this research was to investigate the effect of sorbitol on the quality ofbacterial cellulose-based biodegradable plastic from pineapple skin waste. Various concentrations of sorbitol as the plasticizer were added into a fermentation medium in the synthesis of bacterial cellulose-sorbitolcomposite (BCSC). The BCSCobtainedwas purified by soaking it into a cycled water-NaOH-water. The purified BCSC wasthen compressed using a hot press with the pressure of 300 psi to obtain BCS plastic. The BCS plasticwas characterized in the aspect of physical properties (water content,the degree of swelling), mechanical properties (tensile strength, elongation, elasticity), molecular structure (functional groups, thedegree of crystallinity), and biodegradability. The results showed that the percentage ofwater content anddegree of swelling of the BCS plastic increased with the addition of sorbitol concentrations. The maximum tensile strength was obtained with the addition of 10.5% sorbitol (%v/v). The degrability of the BCS plastic was up to 59% until the 9th day. TheFTIR results showed that functional groups appeared in the BCS plastic were similar to those of the BC. The degree of crystallinity of the BCSplastic decreased with the increasing concentration of sorbitol.

Keywords: Pineapple skin extract, bacterial cellulose, sorbitol, biodegradable plastic, composites

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1 Introduction

Plastic is a material which is the result of the polymerization reaction from similar monomers which form elastic long chains. Plastic utilized as food packaging derived from the processed results of polymerized petroleum and natural gas. In addition to the nature of plastic as non-biodegradable, plastic materials also contain additives that can be absorbed into the packed food ingredientsso itwill endanger the consumers' health (Sopyan, 2001). One of the solutions to reduce the problems posed by plastic is by developing biodegradable plastic which is harmless to food. Biodegradable plastic is plastic which is easily degraded (Jannah, et al., 2014).

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Bacterial Cellulose (BC) is straight-chain polysaccharides composed of D-glucose molecules through β-1,4 glycoside bonds. Studies show that BC is chemically identical to plant cellulose, but the characteristics are different from cellulose based on the macromolecular structure. BC which is produced from the fermentation process has better physical charactersthan plant cellulose, such as high water absorption, high crystallinity, great elasticity, excellent biocompatibility, and high purity.

Pineapple is one of the most favorite fruits because it contains manycarbohydrates and other nutritional sources. However, most people are not aware of the waste produced. The pineapple waste which can be produced is including wet skin and its humpwhich will rot and produce an unpleasant odor in a short amount of time and might disrupt the surrounding ecosystem (Bartholomew, et al. 2003). As pineapple skin contains relatively high carbohydrate and sugar, the skin might be used as a raw material for the production of biodegradable plastic with the fermentation method of BC materials.

BC-based plastic has a rigid and less plasticity nature.An additional plasticizer is required to obtain more plasticity in the plastic. One of the plasticizers is sorbitol. A plasticizer is an organic material withlow molecular weight, non-volatile nature, and high boiling point. Furthermore, it can also change the characteristics of a material. The addition of plasticizer can weaken the strength of polymer and increase the flexibility and extensibility of polymer. A plasticizer is generally classified based on the chemical composition as it will be easier to understand the effect of the element structure on the plasticizer properties and its impact on the materials containing a plasticizer in it (Wipich, 2004).

The addition of plasticizer can reduce the fragility of the plastic films and increase flexibility. Different types and concentrations of plasticizer might affect the film thickness and flexibility, density, and water content (Senyang, et al., 2015). Furthermore, Jannah and colleagues(2014) have investigated the effect of adding corn sugar on the characterization and degradation of biodegradable plastic from cassava starch water. The results showed that the addition of corn sugar influenced the flexural strength of the plastic produced. In other words, if more corn sugar was added, the flexural strength will also increase.

One of the plasticizers which can be used is sorbitol. Sorbitol has a similar structure to glucose so thata strong intermolecular interaction between sorbitol and polymers occurs (Senyang et al., 2015). Furthermore, sorbitol is a polyhydric alcohol monosaccharide compound. Another chemical name of sorbitol is hexitol or glucitol, andits chemical formulais $C_6H_{14}O_6$. The molecular structure of sorbitol is similar to the molecular structure of glucose. The only difference isthat the aldehyde group in the glucose is replaced with an alcohol group.

This study aimed to determine the effect and characteristics of the addition of sorbitol on the quality of BC-based biodegradable plastic from pineapple skin waste.

2 Materials and Methods

2.1 Tools and Materials

The tools used in the production and characterization in this study were glassware, plastic containers, cooking pot, stove, duster, newspapers, tissue rolls, filter, rubber, stirrer, knife, scissor, pH paper, technical balance, analytical balance, heat press, desiccator, evaporator cup, Iron (Myako), oven, tensile strength tool (1990 Tension Testing ASTM vol. 03.10), Fourier Transform Infra Red (FTIR) from Perkin Elmer, and X-Ray Diffraction (XRD). In addition, the materials used in the study were pineapple skin waste, A. xylinum inoculum (from the Laboratory of Biochemistry, UNP), sugar, 25% vinegar, urea fertilizer, aquadest, 70% sorbitol (Bratachem), and NaOH (Bratachem).

2.2 Preparation of the Pineapple Skin Extract

The pineapple skin extract was obtained by smoothing and separating the filtrate from the dregs through filtration. The solvent used to extract was water.

2.3 Production of Bacterial Cellulose-Sorbitol

The production of Bacterial Cellulose-Sorbitol (BCS) was done by inserting the pineapple skin extract into a cooking pot, and then a few grams of sugar, urea, and mL of acetic acid were added into the pot. Afterward, sorbitol was added with some variations (0%, 3.5%, 7%, 10.5%, 14%). All ingredients were heated to boil. The boiled mixture was poured into a plastic container with a height of ±1 cm each and was closed with sterilized newspapers. After the mixture was left and reached room temperature, A. xylinum bacteria was put into the material. The fermentation was done until BCS sheets were formed. The sheets were washed with running water for 24 hours, and then the process was continued by soaking it into NaOH for 24 hours. Finally, the sheets were washed again with clean water.

After BCS was clean, it was pressed using a hot press with a pressure of 300 psi and was dried through heating. Then, the plastic sheets were prepared to be measured by its physical properties, mechanical properties, degradation ability, molecular structure, and degree of crystallinity.

2.4 Water Content Test

Water content is the comparison of the BCS weight before it is pressed and the BCS weight after it becomes a plastic sheet. The percentage of water content was calculated by the following equation:

$$\% \text{ Water Content} = \frac{\text{wet weight-dry weight}}{\text{wet weight}} \text{x} 100\%$$

2.5 Degree of Swelling

The degree of swelling was conducted to know the plastic ability to swell again. The degree of swelling is the comparison of the BCS plastic weight after submersion (constant) and the weight of the dry BCS plastic. The percentage of the degree of swelling was calculated by the equation:

$$\% \text{ Degree of Swelling} = \frac{\text{constant weight-initial weight}}{\text{initial weight}} \text{x}100\%$$

2.6 Tensile Strength Analysis

Tensile strength is the ability of a material to withstand the pull until it breaks. The tensile strength of the BCS plastic was tested using a tensile test tool.

2.7 Elongation Analysis

Elongation is the percentage of the material's ability to elongate. The percentage of elongation was obtained by comparing the length increase of the plastic material to the initial plastic length.

2.8 Biodegradation Test

Biodegradation test was conducted to examine the ability of plastic to be decomposed by microbes in the soil. In other words, the easier the plastic to be decomposed, the quality of the plastic produced will be better. The BCS plastic was degraded for 18 days at a soil depth of 15 cm by controlling the soil conditions used. The percentage of Mass Loss (ML) was calculated by using the following equation:

$$\% \text{ ML} = \frac{\text{initial weight} - \text{final weight constant weight-initial weight}}{\text{initial weight}} \text{x}100\%$$

3 Results and Discussion

3.1 Bacterial Cellulose-Sorbitol

Bacterial cellulose-sorbitol (BCS) is the conversion result of glucose to cellulose with the addition of sorbitol as a plasticizer. Different treatments with the addition of plasticizer in the fermentation medium will produce different cellulose. This difference can be seen in Table 1.

Table 1 shows the effect of adding sorbitol to the BCS fermentation time in which the greater the concentration of sorbitol added, the longer the fermentation time is required. In addition to being a plasticizing agent, sorbitol also acts as an inhibitory agent which can inhibit the rate of fermentation reaction.

This is consistent with aprevious study about the addition of nutrients which exceeded the optimum concentration for the growth of A. xylinum. Converting glucose to cellulose causes plasmolysis (dehydration) in A. xylinum cells, and it can reduce the formation of cellulose (Iskandar, et al., 2010).

Table 1. Treatments of the BCS Formation

Treatment	0% BCS	3.5% BCS	7% BCS	10.5% BCS	14% BCS
Number of medium	400 ml	400 ml	400 ml	400 ml	400 ml
Number of *plasticizer*	0 ml	20 ml	40 ml	60 ml	80 ml
Number of starter	40 ml	40 ml	40 ml	40 ml	40 ml
Tray size	24 x 17 x 4 cm	24 x 17 x 4 cm	24 x 17 x 4 cm	24 x 17 x 4 cm	24 x 17 x 4 cm
Duration of Fermentation	7 days	7 days	7 days	7 days	7 days
Thickness	5.22–6.27 mm	7.27-8.52 mm	3.87-4.01 mm	2.54-2.98 mm	0.98-1.34 mm
The form of cellulose	Thick, solid, and difficult to elongate	Thick, solid, and easy to elongate	Not too thick and soft	Thin and soft	Very thin and soft
Duration of Fermentation	7 days	7 days	10 days	14 days	14 days
Thickness	5.22-6.27 mm	7.27-8.52 mm	6.22-6.34 mm	8.31-9.22 mm	7.23-8.27 mm
The form of cellulose	Thick and difficult to elongate	Thick and easy to elongate	Elastic and easy to elongate	Elasticand easy to elongate	Very elastic and easy to elongate

3.2 BCS Plastic

The drying of BCS plastic was done through 3 ways, namely drying using an oven at a temperature of 1250C for 10 minutes, drying using solar thermal for 1 hour (12.00-13.00 WIB), and drying using an iron (Miyako) with maximum heating. The results obtained are shown in Figure 1.

Comparison of the three samples showed that BCS plastic which did not shrink, was not easy, and the surface was slippery was by drying using an iron. Thus, this plastic was used for further characterization.

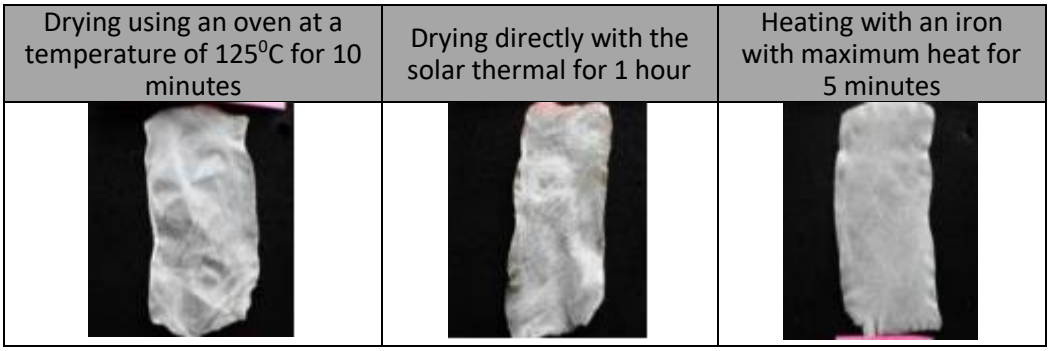

Figure 1.Comparison of the BCS Plastic with several drying techniques

3.3 Water Content in BCS

Water content is the total amount of water contained in a material divided by the dry weight of the material. Water content is a test parameter of physical properties to determine the amount of water contained in a BCS plastic formed from fermentation. The effect of adding sorbitol in the BCS plastic preparation medium to the percentage of water contained in cellulose can be seen in Figure 2.

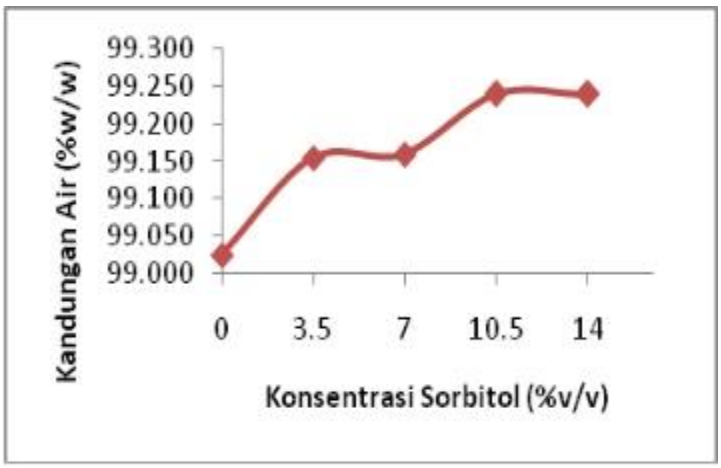

Figure 2.The effect of adding sorbitol on the percentage of water content in BCS plastic.

Based on Figure 2, the percentage of water contained in 0% BCS was ± 99.025%. In other words, the percentage of water content increased with the addition of sorbitol as the plasticizer. The percentage of water content in BCS reached the optimum state (± 99.240%) with the addition of 10.5% sorbitol. This data supported the results obtained in Table 1. Based on the previous research, A. xylinum can convert sugar well which leads to the reduced water content in BCS (Hermawan, 2004). This finding was consistent with the results obtained in this study that 0% BCS had a lower water content than 3.5%, 7%, 10.5%, and 14% BCS. It showed that the presence of sorbitol in the fermentation medium resulted in the decreasing ability of A. xylinum to produce cellulose. Moreover, sorbitol has a hydrophilic nature which can bind water. Thus, the BCS produced had a greater water content than the BC.

3.4 Degree of Swelling

The degree of swelling is one of the physical parameters of the BCS test which shows the ability of BCS cellulose plastic to be swelled again (Megasari, 2015). The effect of adding sorbitol on the degree of swelling percentage of the BCS plastic was weighed every 24 hours until constant at room temperature.

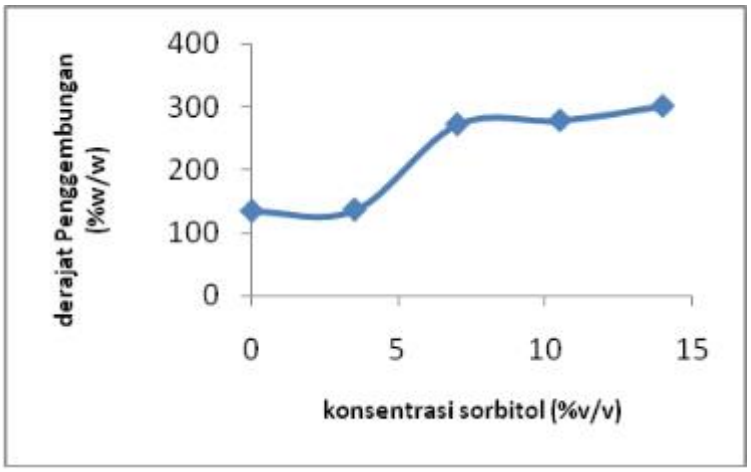

Figure 3.The effect of adding sorbitol on the degree of swelling percentage of the BCS plastic.

As seen in Figure 3, the effect of adding sorbitol in the BCS plastic preparation medium was largebecause it could increase the degree of swelling percentage. The degree of swelling percentage of the 0% BCS plastic was ± 135% whereas the degree of swelling of the 3.5% BCS plastic had a slight increase to ± 137%. The degree of swelling increased sharply in the 7% BCS plastic with ± 273%. The increase in the percentage of sorbitol added also resulted in an increase in the percentage of the degree of swelling. This is consistent with the increasing percentage of water content with the increasing concentration of sorbitol added (Figure 2).

The degree of swelling percentage of the BCS plastic with the addition of sorbitol plasticizer was higher than that of the 0% BCS plastic (without the addition of sorbitol). This is because the addition of sorbitol caused a higher number of O-H groups contained in the BCS plastic than the BC plastic. As a result, the BCS plastic can bind more water molecules which lead to the increased degree of swelling. Moreover, the BCS plastic may also have less dense pores because the degree of swelling percentage is influenced by the pore density and the presence of cross-links which connects two main bacterial cellulose-forming polymers (Putra et al., 2008). The less dense the pores of the BCS plastic, the easier it is to inflate the plastic.

3.5 Tensile Strength

Tensile strength is an important analysis in making plastics because tensile strength determines the quality of the plastic. The effect of adding sorbitol on theBCS plastic is shown in Figure 4. Figure 4 showed that the 0% BCS plastic had a tensile strength of 2.58 kN/m. The tensile strength value of plastic increased along with the addition of sorbitol to the BC fermentation medium. The largest tensile strength value was shown by the 10.5% BCS plastic. However, the tensile strength value of BCS plastic decreased with the addition of 14% sorbitol. Thus, it can be concluded that BCS plastic had maximum tensile strength values if sorbitol was added up to 10.5%.

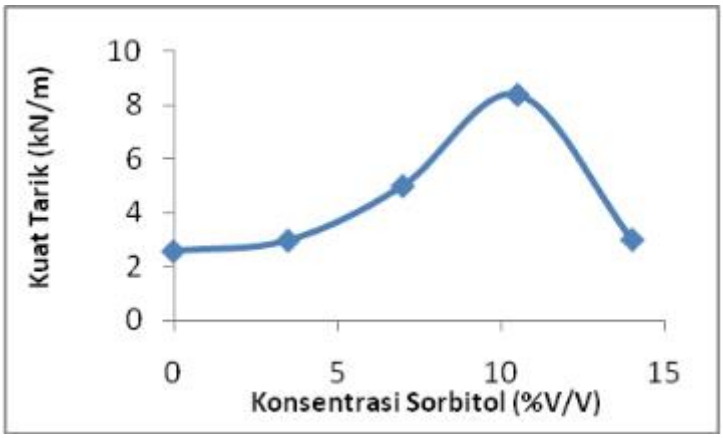

Figure 4.The effect of adding sorbitol on the tensile strength of the BCS plastic.

This result is consistent with a previous study about cassava starch extract (nata plastic) using corn sugar which found that tensile strength of biodegradable plastics increased with the increasing concentration of corn sugar up to 9g. The addition of corn sugar as a plasticizer can bind particles to the empty space contained in the plastic pores (Jannah, et al., 2014). Thus, the plastic gives a large force in the tensile strength test. In this study, the addition of sorbitol had a maximum limit of up to 10.5%. The addition of sorbitol by more than 10.5% will decrease the tensile strength of the BCS plastic. If compared with the results of previous studies, the tensile strength of the BCS plastic was greater with 8.4 kN/m while the plastic produced was 0.13 kN/m (Jannah, etal., 2014). This finding means that the tensile strength value of the BCS plastic is better.

3.6 Elongation of BCS Plastic

Elongation is a mechanical property which is closely related to the mechanical properties of plastic. Elongation shows the maximum plastic length change when receiving tensile strength until the plastic breaks. Elongation values of bacterial cellulose-sorbitol plastic are shown in Figure 5.

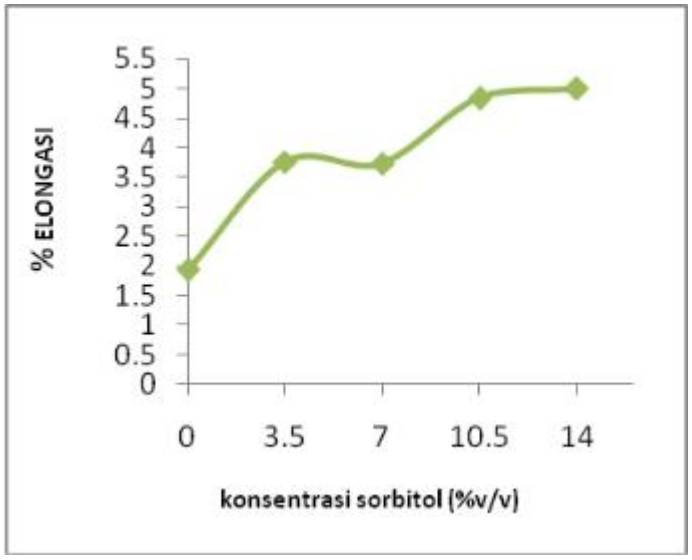

Figure 5.The effect of adding sorbitol on the percentage of elongation of the BCS plastic.

As seen in Figure 5, the ability of plastic continues to increase when the concentration of sorbitol increases. The optimum condition was found in the BCS plastic when 10.5% sorbitol was added, but there was no increase in the addition of 14% sorbitol. However, based on Figure 4, the tensile strength of plastic had a drastic decrease with the addition of 14% sorbitol. This means that the more sorbitol is added,the more elastic the plastic produced, but the tensile strength is lower. Thus, the produced BCS plastic would have good elasticity and maximum tensile strength in the variation of 10.5% sorbitol added. This is consistent with a previous study which found that the addition of sorbitol as a plasticizer was directly proportional to the percentage of elongation (strain). In other words, if more sorbitol was added, the percentage of elongation/strain values was also greater (Senyang, et al., 2015).

Furthermore, plastic elongation also depends on the thickness of the plastic produced and the cellulose fibers produced from the fermentation of cellulose formation by A. xylinum (Jannah et.al., 2014).The physical properties of cellulose derived from the results of glucose fermentation by bacteria had high mechanical strength and porosity, had large water absorption capacity, and easily decomposed (Ksrystynowichz, 2001).

3.7 Modulus Young of BCS Plastic

Modulus young value determines the elasticity value of BCS plastic. Modulus young can be determined through a comparison between the tensile strength value (Stress) and the elongation at the time of breaking (Strain).

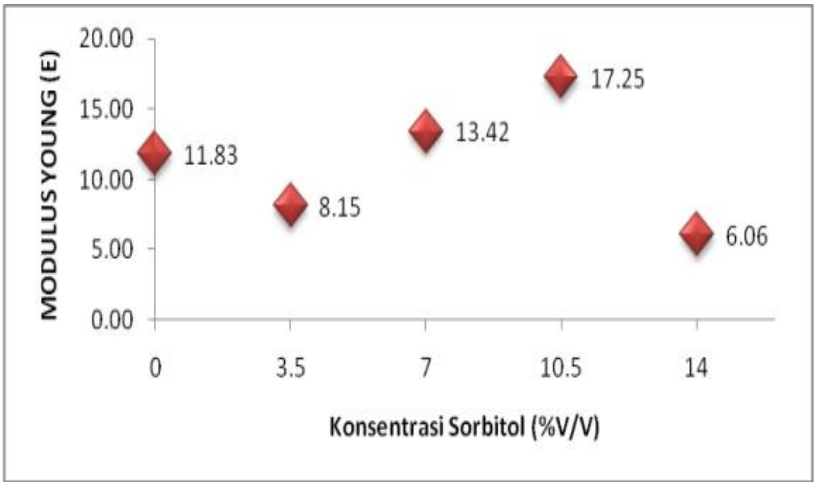

Figure 6.The effect of adding sorbitol on modulus young value (elasticity) of the BCS plastic.

Based on Figure 6, the modulus young value of 0% BCS plastic was 11.83 kN/m, and the maximum modulus young value of the BCS plastic was 17.25 kN/m seen in the addition of 10.5% sorbitol. Thus, the 10.5% BCS plastic had a greater level of elasticity than the 0% BCS plastic.

3.8 Degradation Ability of BCS Plastic

Biodegradation test was conducted to determine the plastic resistance produced against decomposing microbes, soil moisture, soil pH, and soil temperature. Biodegradation test was done by examining the effect of burial time on the percentage of sample mass loss and the surface analysis of samples before and after the burial. The soil conditions in this study were controlled including pH, humidity, and temperature. The occurrence of biodegradation was characterized by the termination of the polymer chain shown by weight loss (Rohaeti, 2009). The ability of the BCS plastic to be degraded by microbes in the soil is presented in Figure 7.

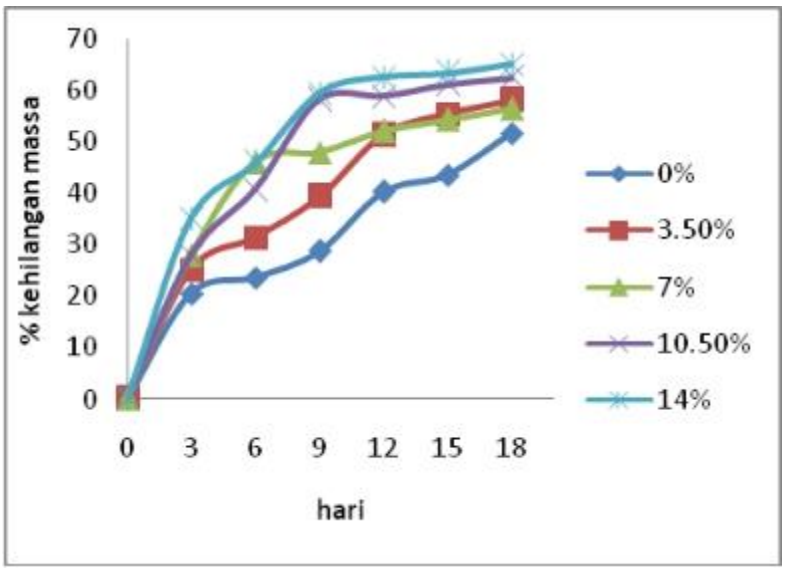

Figure 7.The effect of adding sorbitol on the percentage of mass loss of the BCS plastic.

In Figure 7, the BCS plastic can be degraded up to 60% for 18 days. The BCS plastic will be more easily degraded with the addition of sorbitol than the BCS plastic without the addition of sorbitol. In other words, the more sorbitol plasticizer is added, the easier the BCS plastic to degrade. The highest degradation ability occurred in BCS plastic with the addition of 14% sorbitol which was 65.18% up to the 18thday.

Plastic biodegradation analysis was also done visually. Figure 8 shows the plastic appearance on day-0 which looked 100% intact and has not experienced any visual damage. On the 9th day of burial, the plastic had a slight change, such as there were small holes in the plastic surface. The plastic continued to experience changes in the structure as holes and pores became increasingly enlarged due to microbes.It showed that the addition of sorbitol as a plasticizer caused the formed plastic to be favored by microbes so that it was easily degraded. Polymer degradation is used to express physical changes due to chemical reactions that include breaking bonds in the backbone of the macromolecules. Chemical degradation reactions in linear polymers cause a decrease in molecular weight or shortening of the chain length (Surdia, 2000).

Figure 8.Differences in the bacterial cellulose-sorbitol plastic before (day-0) and after degraded (day-9).

3.9 Analysis of the Bacterial Cellulose-Sorbitol Functional Groups

Functional group analysis was used to determine the difference in functional groups between 0% BCS plastic and 10.5% BCS plastic. The functional group analysis was done using an FTIR spectrophotometer. FTIR spectrum results wereanalyzed qualitatively to find the functional groups contained in each 0% BCS plastic and 10.5% BCS plastic. Figure 9 shows the FTIR spectrum of 0% BCS plastic and 10.5% BCS plastic.

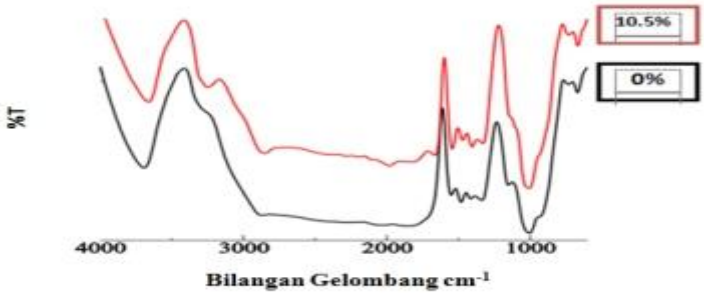

Figure 9. FTIR Spectrum of the BCS Plastic

Based on the FTIR spectrum in Figure 9, the functional groups contained in the BCS plastic can be analyzed. The peaks that appeared are shown in Table 2.

Table 2.The Wave Number of the Peak Spectra of the BCS Plastic

Peak (cm^{-1})	0% BCS (A1)	10.5% BCS (A4)
O-H	3693.67 cm^{-1}	3691.50 cm^{-1}
C-H	2872.15 cm^{-1}	2873.54 cm^{-1}
-C-H	1411.69 cm^{-1}	1410.63 cm^{-1}
C-O	1008.43 cm^{-1}	1007.61 cm^{-1}
-C-O-C	1149.26 cm^{-1}	1148.54 cm^{-1}

Based on Figure 9, the plastic produced from the fermentation by A.xylinum was aBC plastic because the functional groups seen in the FTIR spectrum were functional groups owned by BC. In the spectra, there was a vibration at the wave numbers of 3100–3700cm^{-1}which showed a gap of O-H alcohol, and the peak at these wavelengths for the 0% BCS plastic had a stronger intensity than the 10.5% BCS plastic. This is due to differences in hydrogen bonds occurred because of different treatment in the preparation of the fermentation medium. The addition of sorbitol to the BCS fermentation medium did not cause the emergence of new peaks. This indicates the absence of new functional groups due to the interaction of cellulose with sorbitol plasticizer (Yusmarini, 2004).

3.10 Degree of Crystallinity of the Bacterial Cellulose-Sorbitol Plastic

Crystallinity test was conducted to determine the degree of plastic crystallinity using an X-ray Diffractogram (XRD). The BCS plastic sample produced had a semicrystalline characteristic which means that it had amorphous or crystalline parts. Plastics that have crystalline parts have sharp peaks whereas amorphous structures produce wide peaks. The structure of commercial plastics is generally semicrystalline which consists of crystalline and amorphous. In other words, whenthe plastic has more crystalline structure, the plastic will be stronger. In addition, when the plastic has more amorphous structure, then the plastic becomes more flexible. The plastic tested on the crystallinity test was 0% BCS plastic and 10.5% BCS plastic.

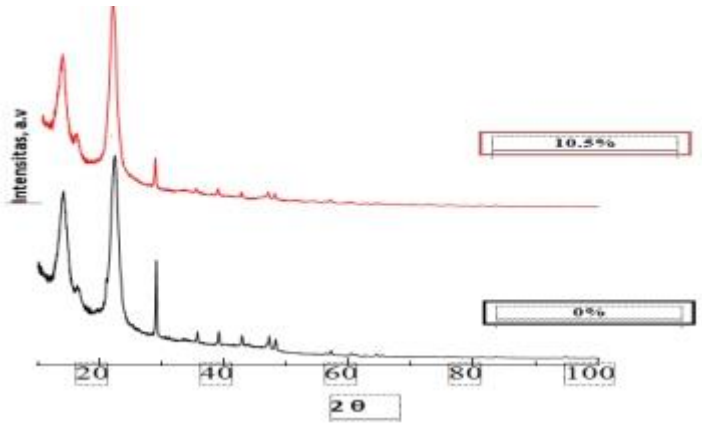

Figure 10. Diffractogram of the BC Plastic (0% v/v) andthe BCS Plastic (10.5%v/v)

Figure 10 shows the diffractogram of the 0% BCS (only BC) and the 10.5% BCS plastic. The resulting diffractogram shows that the cellulose produced was cellulose I. This is shown from the peaks that appeared around 2θ 14o, 16o, 22o dan 22.48o (Yue, etal., 2013). Based on the BCS diffractogram, the 0% BCS plastic had a sharper and more intense peak than the 10.5% BCS plastic.

The degree of crystallinity can be determined manually using a method based on the comparison between the weight of the crystal and the overall weight (amorphous weight + crystalline weight) (Terinte, et al., 2011).

Table 3. The Percentage of the Crystallinity Degree of the BCS Plastic

Sample	Total Weight (g)	Amorphous Weight (g)	Crystalline Weight (g)	% Degree of Crystallinity
BCS 0%	0.1501	0.12	0.0301	79.94 %
BCS 10.5%	0.1354	0.105	0.0304	77.54 %

Based on the calculations that have been made, the degree of crystallinity of the 0% BCS plastic was 79.94% which means that the 0% BCS plastic had 20.06% of the amorphous structure. On the other hand, the degree of crystallinity of the 10.5% BCS plastic was 77.54% which means that it had 22.46% of the amorphous structure. Based on the percentage of the amorphous structure, the 10% BCS plastic was more amorphous than the 0% BCS plastic. This is consistent with the tensile strength values of both samples in which the tensile strength value of the 10.5% BCS plastic was greater than that of the 0% BCS plastic because the 0% BCS plastic had a more crystalline and rigid structure. Thus, sorbitol can reduce the degree of crystallinity of the BCS plastic.

4 Conclusion

In conclusion, the addition of sorbitol as a plasticizer influenced the fermentation time of BCS. The percentage of water content, the degree of swelling, degradation ability increased with the addition of greater sorbitol concentration. The maximum tensile strength was obtained in the 10.5% BCS plastic. The percentage of plastic elongation increased with the increasing amount of sorbitol added. The highest modulus young was found in the 10.5% BCS plastic. The FTIR spectra showed the presence of cellulose functional groups, but it did not show the presence of new functional groups in the plastic with the addition of sorbitol. The degree of crystallinity of the BCS plastic decreased in the presence of sorbitol.

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