Journal of Vocational Studies on Applied Research

Characteristics of Biodegradable Plastic from Cassava Starch with α-Cyclodextrin Based on Chitosan and Glycerol Composition

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Abstract – *Plastic waste is one of the big problems in Indonesia. Plastic cannot be decomposed and is degraded by microorganisms. Therefore, research is needed on plastic materials that can be broken down, which is biodegradable plastic. Biodegradable plastic can be made from tuber materials such as cassava starch and chitosan composites, which are easily degraded by microorganisms. The addition of α-cyclodextrin and glycerol is expected to add flexibility and elasticity to biodegradable plastic. At the stage of making biodegradable plastic, biodegradable plastic material is made by using a blending method and dried at an oven temperature of 50°C for 20 hours. The main ingredients of biodegradable plastic are cassava starch 0.65 gram and α-cyclodextrin 3.25 gram. Additional ingredients are glycerol (1; 1.5; and 2) mL and chitosan (1.3; 1.35; and 1.4) grams. The characteristics of biodegradable plastic were carried out by FTIR analysis, tensile strength test, elongation test, and biodegradable test. Biodegradable plastic test results for optimal tensile strength values for 1 mL glycerol and 1.4 grams of chitosan were found to be 5.26 MPa. The optimal percent elongation test results were 131, 7% for 1,5 mL glycerol and 1.3 grams of chitosan. In the biodegradation test, biodegradable plastic was degraded in 7 days.*

Keywords: *biodegradable plastic, cassava starch, alpha-cyclodextrin, chitosan, glycerol*

Received: November 22, 2024 Revised: December 17, 2024 Accepted: December 26, 2024

Doi: http://dx.doi.org/10.14710/jvsar.v6i2.25114

[How to cite this article: Purwanti, O.T., Shabrina, A.N., Dermawan, B., Insani, R.K. (2024). Characteristics of Biodegradable Plastic from Cassava Starch with α-Cyclodextrin Based on Chitosan and Glycerol Composition. Journal of Vocational Studies on Applied Research, 6(2), 34-37. doi: http://dx.doi.org/10.14710/jvsar. v6i2.25114]

INTRODUCTION

Plastic is the largest contributor to waste in Indonesia and the second in the world after China. Based on data from the National Waste Management Information System (SIPSN), the amount of waste piles in Indonesia nationally consisting of 303 regencies/cities is 35.93 tons in 2022 and this amount is predicted to continue to increase. Plastic is made from synthetic materials called polymers. The decomposition of plastic by bacteria takes a very long time or can be said to be non-biodegradable naturally.

Biodegradable plastic is a solution to the above problems. Biodegradable plastic or often called bioplastic is plastic that can be broken down by the activity of microorganisms with the end result being water and carbon dioxide gas. However, it can be used like conventional plastic. Biodegradable plastic is generally made from renewable materials, such as compounds found in plants (cellulose, starch, collagen, casein, protein) and animals (lipids).

One of the natural polymers that can be used to make biodegradable plastic is starch. Starch ($C_6H_{10}O_5$) is a carbohydrate compound of the polysaccharide group from plant extraction. One of the natural ingredients that has a high starch content and can be used in making biodegradable plastic is cassava.

However, biodegradable plastic from starch has weak mechanical properties. Therefore, it is necessary to add plasticizers such as glycerol. The addition of glycerol plasticizer under certain conditions can improve the mechanical and physical properties of biodegradable plastic, such as avoiding tearing, and producing strong and flexible plastic (Kumoro et al., 2014).

In addition, the basic material of biodegradable plastic also requires other chemicals, such as chitosan. Chitosan is a protein modification of chitin that can be found in the shells of shrimp, crabs, lobsters, and insects. Chitosan can improve the transparency and mechanical properties of the resulting plastic. The use of chitosan is also based on its easily degraded nature and easy to combine with other materials. In addition, chitosan is also a preservative from natural materials (Chimie et al., 2016).

Cyclodextrin is the result of various starch degradation which is a natural polymer and can be combined with other types of natural polymers such as chitosan, gelatin, lignin, and polysaccharides. Cyclodextrin will increase stability and other properties, Cyclodextrin with chitosan can help improve antibacterial properties that stabilize by maintaining its solubility (Sun et al., 2014).

This study aims to determine the effect of chitosan and glycerol composition on the characteristics of biodegradable plastic, the mechanical properties of biodegradable plastic from cassava starch with α-Cyclodextrin, and the degradation ability of biodegradable plastic against glycerol composition.

MATERIALS AND METHOD

General Instructions

This research was conducted at Industrial Chemical Engineering Technology Laboratory, Vocational School Faculty, Diponegoro University Semarang from October until January 2024.

Cassava Starch and Cyclodextrin

Experiment on making biodegradable plastic based on cassava starch and cyclodextrin with the addition of chitosan and glycerol. Detailed experimental conditions are given in Table 1. The experimental procedure carried out was making inclusions of starch and α-Cyclodextrin by making a ratio of starch and α-Cyclodextrin of 1:5 where cassava starch was 0.65 grams and α-Cyclodextrin was 3.25 grams. The α-cyclodextrin solution was made by weighing 3.25 grams of α-cyclodextrin powder for each variable dissolved in 50 mL of ethanol solution with a ratio of 70% ethanol: distilled water (1:2, v/v) which was heated to a temperature of 70°C above Hot Plate with stirring at 800 rpm. The α-cyclodextrin solution was cooled to 40°C.

Tabel 1. Experiment Condition

Sample	Raw Material Composition	
	Chitosan (gr)	Glycerol (mL)
A1B1	1	1,3
A1B2	1	1,33
A1B3	1	1,4
A2B1	1,5	1,3
A2B2	1,5	1,35
A2B3	1,5	1,4
A3B1	2	1,3
A3B2	2	1,35
A3B3	2	1,4

Production of Bioplastics

Making chitosan films by dissolving chitosan in 2% acetic acid solution using a magnetic stirrer at a speed of 1000 rpm at a temperature of 70°C for 30 minutes. Then the chitosan solution was filtered with stirring. The resulting solution is clear white and there are air bubbles formed due to stirring. Then the chitosan solution was added to the α-cyclodextrin solution and starch with glycerol was added as a plasticizer. The mixture was heated while stirring using a hot plate and magnetic stirrer at a temperature of 70 °C. Then the cooled mixture is then molded onto a mold, then dried in an oven at 50°C for 20 hours. After the film is dried, the film is left at room temperature and then released from the mold and tested for the physical characteristics of the bioplastic and the biodegradable test.

Bioplastics Analysis

The bioplastic analyses are tensile strength, elongation at break, biodegradability, and Fourier Transform Infrared Spectrophotometry (FTIR) according to ASTM or can be tested in existing laboratories implementing ISO/IEC 17025 (ASTM D882-12, 2002). Bioplastic products compared with bioplastics quality standards according to SNI 7188.7:2016. Tensile strength calculations are carried out to determine the maximum tensile strength or load that can be achieved until the film remains stable before breaking (Lismawati, 2017). The percentage elongation at break can be obtained from the comparison of the increase in the initial length of the sample with the final length of the sample after being pulled to break (Pujawati et al., 2021). Tensile strength and elongation at break can be calculated using equations (1) and (2), respectively.

$$\sigma = \frac{load\ of\ break}{(original\ width)(original\ thickness)} \quad (1)$$

$$\varepsilon = \frac{(the\ final\ length\ of\ the\ test\ object - the\ initial\ length\ of\ the\ test\ object)}{The\ initial\ length\ of\ test\ object} \times 100\ \% \quad (2)$$

Biodegradation by utilizing microorganisms to determine biological reactions. The biodegradation testing method involves placing the sample in a test tube and then pouring in 5 mL of EM4 which has been activated. Then stored for 7 days at room temperature. After 7 days, they were taken and washed with distilled water, rinsed with alcohol and dried in an incubator at 70°C for 24 hours. Then weigh it and calculate the mass reduction. The biodegradation test can be calculated using equation (3).

$$Weight\ Residual(\%) = \frac{(The\ initial\ weight\ before\ biodegradation\ test - the\ final\ weight\ after\ biodegradation\ test)}{The\ initial\ weight\ before\ biodegradation\ test} \times 100\ \% \quad (3)$$

RESULTS AND DISCUSSION

Analysis of Functional Group with FTI

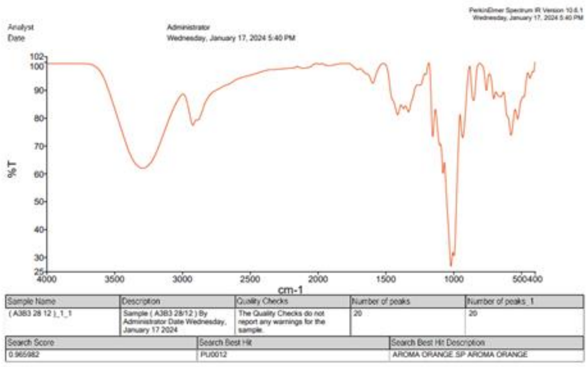

Figure 1. Analysis of Functional Groups of Biodegradable Plastic wit FTIR

Results of the FTIR spectrum of the biodegradable plastic produced. It can be seen that the spectrum of biodegradable plastic produced. It can be seen that the spectrum of biodegradable plastic shows several waves whose values are discussed in Table 1. This shows that the functional groups of the biodegradable plastic produced are almost the same as the functional groups specific for biodegradable plastic. From the FTIR graph there are several absorption peaks, namely the range 3000-3500 cm^{-1} shows the O-H group, and at 2000-3000 cm^{-1} the wave shows the C-H group. In the other hand from that, there is aromatic group C=C (alkene) at 1550 cm^{-1} and a 1550 cm^{-1} and a C-O group (alcohol) at 1006-1300 cm^{-1}. The group context is an organic group, so this biodegradable plastic material is environmentally friendly because it is easily degraded by microorganisms. Inside that, the presence of hydrophilic C-O and O-H groups also makes it possible for them to bind with water, which makes it easier for plastic to degrade more quickly.

Tensile Strength

Figure 2 shows that the tensile strength of plastic increases with the addition of chitosan mass composition. The higher the chitosan mass composition, the more C≡C groups will be contained in the plastic. This causes the intermolecular bonds of the plastic to be stronger and more difficult to break, because it requires a lot of energy to break the bonds (Siro et al., 2006). The addition of chitosan mass composition causes the molecular structure of biodegradable plastic to be denser and more homogeneous, so that the tensile strength value will be higher (Safitri et al., 2016).

Elongation of Break

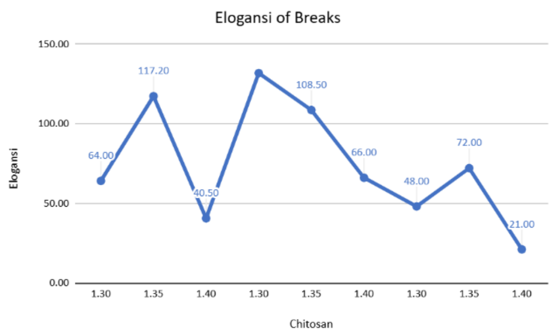

Figure 3. Biodegradable Plastic Elongation of Break Graph

The graph results above show that the elongation value decreases as the chitosan mass composition increases. The elongation value of plastic is inversely proportional to its tensile strength (Sun et al., 2014). Plastics that have the most C≡C groups are difficult to remove so they tend to be difficult to stretch and have a smaller elongation value. This is due to the decreasing bond distance between molecules (Safitri et al., 2016).

Biodegradation Test

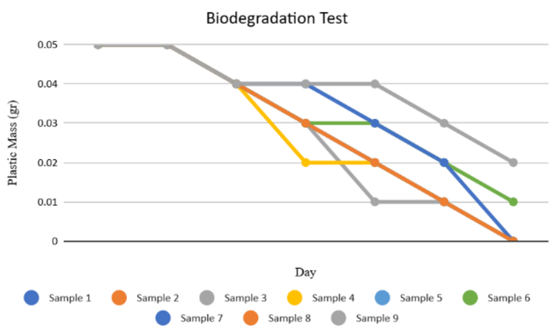

Fig 4. Biodegradable Test Graph

The graph above presents the results of the biodegradation test for 7 days. The higher the proportion of cassava starch, the faster the degradation will be due to its hydrophilic nature. However, in this experiment, the amount of cassava starch in all samples remained at 0.65 grams. In contrast, the amount of chitosan varied, with 1.3, 1.35, and 1.4

grams used. The sample with the highest chitosan content showed stronger molecular bonds, indicated by the high intensity of the C≡C group. This resulted in stronger intermolecular bonds in the plastic, making it more difficult to break down. In addition, the hydrophobic nature of chitosan meant that the sample with the highest mass of chitosan took longer to degrade (Hartatik et al., 2014).

CONCLUSION

Cassava flour and shrimp shell chitosan can be used as basic materials for making biodegradable plastic. The addition of chitosan mass composition has been proven to increase the tensile strength value of biodegradable plastic. Meanwhile, the elongation value is inversely proportional to the tensile strength value. The high intensity of C≡C bonds in samples with the highest chitosan mass composition causes the bonds to be more difficult to stretch/break because they have a high average bond energy, thus increasing the tensile strength value and slowing down the degradation process.

FTIR analysis revealed absorption peaks corresponding to various functional groups, which are characteristic of organic compounds. Specifically, the O-H group was observed at 3000-3500 cm^{-1}, the C-H group at 2000-3000 cm^{-1}, the aromatic group C=C (alkene) at 1550 cm^{-1}, and the C-O group (alcohol) at 1006-1300 cm^{-1}.

The degradation rate of the sample is influenced by the balance between cassava flour which is hydrophilic and accelerates degradation, and chitosan which is hydrophobic and slows degradation due to its strong molecular bonds (high intensity of C≡C groups).

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