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Synthesis of Eco-Friendly Bioplastic from Banana Hump Starch and Sugarcane Bagasse Cellulose Filler with Fish Scale Chitosan

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Abstract. This study aims to make environmentally friendly bioplastics. This is because the use of plastic creates a plastic waste problem which is very difficult to handle. After all, it is difficult to decompose. Excessive use of plastic will be bad for the safety of the earth in the future. One of the innovations to be developed is the synthesis of environmentally friendly bioplastics. The basic ingredients for making bioplastics come from starch, cellulose, and chitosan. The method for making bioplastics begins with sample preparation on banana stems, sugarcane bagasse cellulose and fish scale chitosan. The process of making bioplastics uses several formulations including A0 (control), A1, A2 and A3. The research results showed that the biodegradability test was good for the A3 formula and the water absorption test was 3.77% and water resistance was 96.2%.

Introduction

The use of plastic often causes waste problems, namely plastic waste, because it is used excessively and is difficult for nature to decompose naturally (Radtra AHA *et al.*, 2021). One innovation that can be made to reduce the use of plastics made from synthetic polymers is making plastics made from natural polymers, which are usually called bioplastics. Bioplastics are plastics made from materials that can be broken down by microorganisms and do not leave toxins (Radhiyatullah *et al.*,2015).

Starch is a carbohydrate contained in plants that is widely and widely found in nature. Starch is not only found in plant fruit but can also be obtained from plant seeds and stems. Therefore, it is necessary to develop bioplastic raw materials from starch waste that are economically valuable and environmentally friendly (Ramadhan *et al.*,2021).

Banana hump is one of the banana wastes that is underutilized. This shows that banana tuber starch can be used as a basis for bioplastics. Bioplastic synthesis requires additional filler to increase tensile strength. The fillers used in making bioplastics can be inorganic and organic fillers. Inorganic fillers such as clay, CaCO3 and ZnO.

Meanwhile, the organic filler is cellulose from natural materials (Hutabalian et al.,2020). The addition of cellulose filler to bioplastic synthesis shows better results by increasing the physical and chemical properties of the biofilm (Effendi DB *et al.*,2015). Naturally abundant cellulose can be obtained from agricultural waste biomass, one of which is from sugarcane bagasse waste. The cellulose content of bagasse is 30.4%. This is a reference that bagasse has the potential as a bioplastic filler (Rahyani, 2011). To provide greater strength attraction to bioplastics, it is necessary to add chitosan as reinforcement and water resistance of bioplastics (Marbun and Eldo S, 2012).

Chitosan is often obtained from the isolation of chitin which comes from the shells of crustaceans such as crabs, shrimp, clams (Aziz *et al.*,2017). In addition, chitosan can also be found in fish scales. Fish scales are the result of waste that is simply thrown away. Accumulated fish scale waste will cause an unpleasant odor and disturb the beauty of the environment.

It is hoped that the results of this research will produce products that have sales value, are economical and environmentally friendly. Reducing plastic waste is one of the actions of going green so this research will help with actions to save the earth through the use of bioplastics by society in the long term.

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Experimental

Material and Methods

The research materials used were banana hump, sugar cane bagasse, fish scales were taken from the Bulukumba Regency area, South Sulawesi, hydrochloric acid (HCl), sodium hydroxide (NaOH), aquadest (H_2O), sorbitol rom the Muhhadiyah Bulukumba University laboratory.

This research will go through several stages starting from preparation, isolation, bleaching, bioplastic synthesis, and bioplastic characterization.

Procedures

Analysis of Water and Ash Content

The water content test was carried out by: evaporating a cup that had been dried and whose weight was known, filled in as much as 2 grams of sample and then weighed (W1) and then put in the oven at 105 °C for 1-2 hours. The evaporating cup and the dried sample are put into the desiccator and then weighed. Heating the sample was repeated until a constant weight (W2) was obtained. The rest of the sample is calculated as total solids and lost water as water content (Nofiandi, 2019).

The ash content test was carried out by: 2 grams of sample was weighed in a porcelain cup of known weight (A), then charred using a Bunsen heater until no more smoke was emitted. A porcelain cup containing sample (B) which has been charred is then put into a 600 °C furnace for 2 hours to turn it into ashes (C). The porcelain cup containing the ash is cooled in a desiccator and weighed until it reaches a constant weight (Nofiandi, 2019).

Bioplastic Synthesis of banana hump starch, sugarcane bagasse cellulose and fish scale chitosan

Bioplastic Synthesisare made by formulating banana hump starch, sugarcane bagasse cellulose and fish scale chitosan. The formula for making biologics can be seen in Table 1.

Table 1. Bioplastic formulations

Bioplastic	starch (%)	cellulose (%)	chitosan (%)
A0	-	-	-
A1	6	2%	2%
A2	5	2,5%	2,5%
A2	4	3%	3%

Stir the bioplastic formula using a magnetic stirrer at 240 rpm until a homogeneous solution is formed. The solution was poured onto a 20 × 15 glass plate. After that, it was baked for 5 hours at 70 °C.

Biodegradability Analysis

Bioplastic samples A1, A2 and A3 were put into a test tube, then EM4 was added and then incubated. Then the test tube is observed.

Analysis of Absorption Capacity on Water Resistance

Fill the beaker with distilled water, then cut the bioplastic A1, A2, and A3 to a size of 3×3 cm. Weigh the initial weight of the bioplastic, then put the edible film into a beaker containing distilled water for 30 minutes. After 30 minutes, the bioplastic was removed from the container containing the distilled water and used a tissue on each surface. Then the weight of the sample (W) which has been immersed in the container is weighed (Darni, et al, 2014).

$$\%\ \text{Water Absorption} = \frac{W - Wo}{Wo} \quad (1)$$

$$\%\ \text{Water Resistance} = 100\% - \%\ \text{Absorption Water} \quad (2)$$

Tensile Strength Analysis

The mechanical properties were characterized using a Universal Tensile Testing Machine with a tensile speed of 5 mm/minute, load cell scale of 4% of 100 kgf. Samples A1, A2, and A3 in sheet form were each cut to a length of 130 mm and a width of 8 mm. Testing was carried out using the IK-MT-28-01 test method.

Result and Discussion

Analysis of Water and Ash Content

Analysis of water content and ash content of banana weevil starch, bagasse cellulose and fish scale chitosan was carried out in the laboratory of the Bulukumba Forestry and Environment Agency. The results of the water and ash content analysis can be seen in Table 2.

Table 2. Analysis result of water and ash content

Sample	Water Content (%)	Ash Content (%)
Pati	0.1574	0.0258
Selulosa	0.0090	0.0303
Kitosan	2.2007	2.0468

From the results of the water and ash content analysis carried out, the starch water content was 0.1575% and the ash content was 0.0258%. The cellulose water content is 0.0090% and the ash content is 0.0303%. Meanwhile, for chitosan, the water content obtained was 2.2007% and the ash content was 2.0468%. From the results of the water and ash content obtained, it can be said that the water and ash content of starch, cellulose and chitosan as basic materials for bioplastics does not exceed the 5% limit (Nofiandi, 2019).

Bioplastic Synthesis of Banana Hump Starch, Sugarcane Bagasse Cellulose and Fish Scale Chitosan

In this research, bioplastics were made using the blending method, namely a method of mixing two or more materials into one. The basic ingredients in this research are banana hump starch, sugarcane bagasse cellulose and fish scale chitosan which are used to make bioplastics.

Bioplastics made from starch are generally brittle and stiff, so glycerol needs to be added to increase the flexibility and softness of the polymer material which is a plasticizer. Glycerol is a hydrophilic plasticizer so it is suiTable for hydrophilic film-forming materials such as starch. However, bioplastics from starch and glycerol still have shortcomings regarding bioplastic characteristics. These shortcomings include not being resistant to water and breaking easily, therefore chitosan needs to be added as a reinforcement, water resistance and anti-microbial. In this research, the addition of chitosan aims to bind the hydrogen contained in the plastic so that the chemical bond will be stronger and more difficult to break. Chitosan is dissolved with acetic acid because chitosan can dissolve well using acetic acid solvent. However, bioplastics from a mixture of starch, chitosan and glycerol still have shortcomings regarding the characteristics of bioplastics. These disadvantages include being easily torn, so it is necessary to add cellulose as a chitosan cross-linker. The addition of cellulose aims to bind the chitosan structure and provide a longer shelf life. Then it is homogenized with a magnetic stirrer while heating, which aims to form a homogeneous solution and form a thick solution (Sriwahyuni, 2018).

Biodegradability Analysis

This biodegradability analysis was carried out to determine the resistance of plastic as food packaging from disturbances that can accelerate damage. Plastics made from natural materials generally have a faster rate of damage. One of the causes of plastic damage is the growth of mold. Observations of fungal growth can be seen in Table 3.

Table 3. Analysis result of biodegradability

Day	A0	A1	A2	A3
1	no black spots and dark brown	no black spots and dark brown	no black spots and dark brown	no black spots and dark brown
2	no black spots and dark brown	no black spots and dark brown	no black spots and dark brown	no black spots and dark brown
3	no black spots and dark brown	no black spots and dark brown	no black spots and dark brown	no black spots and dark brown
4	no black spots and dark brown	black spots begin to appear and dark brown	black spots begin to appear and light brown	black spots begin to appear and light brown
5	no black spots and dark brown	dark spots increase and light brown	dark spots increase and light brown	dark spots increase and light brown
6	no black spots and dark brown	dark spots increase and light brown	dark spots increase and light brown	dark spots increase and light brown
7	no black spots and dark brown	dark spots increase and clear brown	dark spots increase and clear brown	dark spots increase and light brown
8	black spots begin to appear and dark brown	decomposed and reddish brown in color	decomposed and reddish brown in color	decomposed and reddish brown in color
9	black spots begin to appear and dark brown	decomposed and reddish brown in color	decomposed and reddish brown in color	decomposed and reddish brown in color
10	black spots begin to appear and dark brown	decomposed and reddish brown in color	decomposed and reddish brown in color	decomposed and reddish brown in color

Biodegradability analysis is carried out to determine the level of resistance of a bioplastic to fungal growth. Plastics made from natural (organic) materials tend to have relatively short durability and shelf life. Bioplastics were previously stored for 10 days to detect the occurrence of fungal growth on the bioplastic layers. The resulting bioplastics were tested for their biodegradable properties using EM4 bacteria (Effective Microorganisms) (Kusumawati Dyah Hayu and Widya Dwi Rukmi Putri, 2013). EM4 is a micro-mixed culture consisting of Lactobacillus, Actinomyces, Streptomyces bacteria, yeast fungi and photogenic bacteria which work to support each other in the decomposition of organic matter. These bacteria will degrade starch-containing bioplastics by breaking polymer chains into monomers through enzymes produced by these bacteria (Ilahand Fina Mahabbatul, 2015).

The results of the biodegradability analysis can be seen that on the first to third day no signs of fungal growth appeared. However, on the fourth day and beyond, the bioplastic samples showed signs of fungal growth on A1, A2 and A3, which was characterized by the appearance of black spots all over the surface of the bioplastic from banana hump starch, sugar cane bagasse cellulose and fish scale chitosan. The slow decomposition of bioplastics is due to the addition of chitosan whose function is to bind hydrogen contained in bioplastics so that the chemical bonds will be stronger and more difficult to break. And the addition of cellulose as a natural fiber filler to bioplastics so that its decomposition is faster than bioplastics without the addition of banana hump starch, sugarcane bagasse cellulose and fish scale chitosan (Mahatmanti, 2010).

From the research results it can be seen that the process of decomposing A1 and A2 is faster than the others. This can be seen on the 7th day which is marked by an increase in black spots and a clear brown color.

Analysis of Absorption Against Water Resistance

Water absorption analysis is carried out to determine the level of resistance of a bioplastic to water. The absorption capacity test is carried out by placing a plastic bioplastic sample into a container filled with water, then the sample is removed and weighed until a constant weight of the bioplastic is obtained. The greater the water absorption, the lower the resistance level of the plastic and the resulting plastic will be damaged quickly (Kaimudin Marni and Maria F. Leounupun). Conversely, the lower the water absorption capacity of a plastic, the greater the resistance level of the plastic and is able to protect packaged products and not be easily damaged/destroyed in water, as can be seen in Table 4.

Table 4. Analysis of absorption against water resistance

Sample	Weight		Water Absorption (%)	water Resistance (%)
	W2	W1		
A0	0.821	1.056	28.6	71.4
A1	0.539	0.656	21.7	78.3
A2	0.331	0.448	11.7	88.3
A3	0.265	0.275	3.77	96.2

From the results of the absorption and water resistance of bioplastics, it can be seen that the absorption capacity of the A3 formula is less and the water resistance is greater than A0, A1 and A2. This is because the amount of cellulose and chitosan in A3 is more than the others. Chitosan and cellulose can increase water resistance and reduce water absorption due to their hydrophobic nature. The greater the amount of chitosan and cellulose, the greater the value of water resistance and lowers the moisture content of bioplastics 90%.

According to the Indonesian national standard (SNI), the water resistance of bioplastics is 99%. In this study, the value that is closest to water resistance based on SNI is bioplastic in formula A3. So this formula is very good for making bioplastic, the higher the water resistance value of the bioplastic, the better the quality of the plastic so that the product to be packaged will last longer. On the other hand, the lower the water resistance of a plastic, the greater the level of damage to the plastic and its solubility in water, so that the packaged product will not last long in storage.

Tensile Strength Analysis

The highest tensile strength results for bioplastics were 0.5109 kgf/cm2 for bioplastics with A3 formulation, while the lowest tensile strength values were 0.3675 for bioplastics with A1 formulation. The tensile strength results obtained were still better than research by Sriwahyuni (2018) with results of 0.0141 kgf/cm2 for corn starch, chitosan, glycerol and glutaraldehyde bioplastics.

The tensile strength value of bioplastic film with the addition of chitosan can be observed that as the cellulose concentration increases, the tensile strength value will increase. This is because the interaction between cellulose and chitosan is getting tighter, causing the resulting bioplastic film to be strong and stiff. During mixing and heating, there is an interaction between chitosan and cellulose to form hydrogen bonds so that the more hydrogen bonds there are in the bioplastic film, which will cause the film to be stronger and more difficult to break (Jannah, 2017).

Table 5. Results Of tensile strength analysis

Parameter Bioplastik	Kuat Tarik (N/mm^2)
A0	0.3675
A1	0.3731
A3	0.3992
A3	0.5109

Based on the data obtained, it can be seen that the tensile strength decreases as fewer components are added. This causes the bonds between molecules in the bioplastic to weaken.

Conclusion

The results showed that bioplastics from banana hump starch, bagasse cellulose and fish scale chitosan had good biodegradability compared to those without banana cob starch, bagasse cellulose and fish scale chitosan. and analysis of water absorption capacity for bioplastic water resistance is the best with formula A3 (4% starch, 3% cellulose and 3% chitosan). From the results it can be concluded that this bioplastic can be categorized as environmentally friendly.

Conflict of Interest

The authors declare that there is no conflict of interest.

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