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Preliminary Properties and Biodegradability of Bioplastics Derived from Mangrove Fruit (*Avicennia lanata*) and Glycerol

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Abstract: Bioplastics are made from starch that can be decomposed naturally to be an alternative to commercial plastics. The research objective was to know the effect of the composition of mangrove powder and glycerol on the characteristics of the synthesized bioplastics. This study also obtained the maximum composition of bioplastics made from mangrove powder by adding glycerol and chitosan. In this study, bioplastics were synthesized using starch as an optimizer of the mechanical properties of bioplastics, chitosan as reinforcement and water repellent, and glycerol as a plasticizer. The results of bioplastics were characterized by mechanical properties (tensile strength and elongation), hydrophobility, biodegradation, and functional groups. Based on the study's results, the optimum composition of bioplastics was found, namely mangrove 2 grams and glycerol 0.9 ml, with a tensile strength value of 1,600 MPa, 67.9% elongation, 34.75% hydrophobility, and 39.98% biodegradation. The main functional groups of bioplastics were shown through the results of FTIR analysis, such as NH (3294.58 cm^{-1}), CH (2883.58 cm^{-1}), OH (2129.41 cm^{-1}), and CO (1755.22 cm^{-1}).

Keywords: bioplastic reinforcement; mangrove powder; glycerol plasticizer; chitosan; enzyme degradation.

1. Introduction

Plastics are widely used in human life, and plastics are now widely used as manufacturing materials that find applications in a wide range of industries, such as packaging, grocery bags, plastic cutlery, etc. Most plastics used are derived from traditional petroleum-based materials [1-4]. Plastics are high molecular weight polymers typically containing 1000 to 10,000 monomeric repeating units [5-7]. So far, many conventional plastics have been used commercially. However, the issue of conventional plastic waste is currently trending due to its environmental impact, including flooding, trash accumulation in landfills, and even air pollution if burned [8]. One way to solve the problem of plastic waste is to create environmentally friendly plastics. Bioplastics are expected to reduce pollution caused by conventional plastic waste, which is extremely hazardous to the environment, and the amount of conventional plastic waste continues to increase over time. Many bioplastics are produced today. Bioplastics are plastics derived from starch-based organic raw materials that are environmentally friendly so that they are easily decomposed by fungi or microorganisms [9-12]. Bioplastics are biodegradable plastics made entirely of natural materials. It can be destroyed in the environment for up to one month in a soil medium [13-17].

Furthermore, bioplastics can decompose into inorganic or organic compounds and biomass through enzymatic degradation by various microorganisms. Bioplastics production is currently restrained because the raw material for making bioplastics is limited and only produced from cassava starch by the Ministry of Industry. It plans to increase production capacity to 500 thousand tons annually but is still constrained by raw materials. To meet these needs, new sources of raw materials are needed. Therefore, many studies have been conducted to produce bioplastics using different technologies [18-22].

According to one of the studies conducted by this reference, biodegradable plastic is based on starch, one of the alternatives to petroleum-based plastic. Because of its abundant availability and high biodegradability, starch is regarded as one of the most promising natural raw materials for producing biodegradable plastic. Starch can be extracted from various sources, including corn, cereal grains, rice, potatoes, etc. Physicochemical properties of biodegradable films made from cassaa, corn, potato, banana peel, microalgae consortium, cocoa pod husk cellulose, jack fruit peel cellulose, and cellulose from jack fruit have become more important in recent years [22-25].

The present study indicates that the raw material for making bioplastics is cellulose, which can be found in plant starch. Therefore, this research will use mangrove fruit as a raw material to manufacture bioplastics. Mangrove fruit has great potential to be used as a basic material for making bioplastics because mangrove fruit has a starch content that is higher than rice, which is 57.73%, consisting of amylose (31.56%) and amylopectin (26.17%). In comparison, rice is 32.91% [26-28]. Hence, biodegradable starch plastics are widely used in packaging applications because of their good barrier properties against oxygen and carbon dioxide [29]. By combining mangrove flour and glycerol, bioplastics were created in this study. Glycerol is used as a plasticizing agent to improve the plasticity of the bioplastics produced.

Plasticized films were more durable and flexible than unplasticized films. A plasticized film with a higher concentration of plasticizer displayed decreased tensile strength and increased elongation at break. As the quantity of plasticizers rose, the water vapor transfer of plasticized films increased, indicating enhanced water permeability but decreased water absorption capacity. Plasticizers considerably decreased biodegradable plastic film swelling and water retention capacity [30]. The study aimed to examine the qualities of bioplastics formed from mangrove fruit, the impact of adding glycerol on bioplastics, the impact of mangrove flour weight on bioplastic properties, and the impact of variations in mangrove flour and glycerol on biodegradability.

2. Materials and Methods

2.1. Materials.

The starch powder was obtained from mangrove fruit. This study uses aqua dest, acetic acid, glycerol, sodium metabisulphite, chitosan, and soil as structural and supporting materials. A total of 1 kg of mangrove fruit was peeled and soaked in water for 3 days before being cleaned with clean water. Mangrove fruit is cut and mashed in a blender with 500 ml of water and 0.05 percent sodium metabisulfite. A calico cloth is used to filter mangrove pulp. Water is added in a 1:1 ratio to the pulp obtained, and the process is repeated twice. For 6 hours, the starch solution was precipitated at 4^{o}C. The starch solution was then deposited for another 12 hours at room temperature until a pure starch powder precipitate was obtained, after which the water was removed. The starch was dried at 60°C for 6 hours, after which the starch flakes were sieved through a 100-mesh sieve to obtain pure starch powder based on mangrove fruit.

2.2. Fabrication of bioplastic films from starch-based mangrove fruit.

Four grams of chitosan were dissolved in 100 mL of 10% acetic acid and stirred with a magnetic stirrer for 30 minutes to make a homogenous slurry. Separate amounts of pure starch powder (0.5 g, 1 g, 1.5 g, 2 g, and 2.5 g) were dissolved in 100 mL distilled water and then heated on a hot plate at 70 °C for 12 minutes while stirring with a magnetic stirrer. Each solution was stirred for 1 hour with glycerol, 15% of the total weight of starch powder, and chitosan until homogenous. The resulting solution was then printed on a plastic mold measuring 20 x 12 cm and 2 cm in height and dried for 6 hours at 60 °C. After cooling the bioplastics for 6 hours at room temperature, they were taken off the glass plate and stabilized in a desiccator before analysis.

2.3. Characterization of bioplastics from mangrove powder.

2.3.1. Tensile test.

A Universal Instron Testing Machine was used to perform the tensile test. The tensile machine, specimen, and conditioning parameters were set up according to ASTM D638. The crosshead was set to move at a speed of 200 mm/min. For each bioplastic composition, an average of three tests was used. Some of the important parameters extracted from the tensile tests were: tensile strength, which was the maximum tensile stress sustained; percent elongation at break, which was the elongation of the sample at the point of rupture; and Young's modulus, which was the stress to strain ratio below the elastic limit.

2.3.2. Water absorption test.

The water absorption of the bioplastic films was determined using modified procedures. All dried films were cut into 6 cm x 2 cm pieces, and the initial weight was recorded. They were then immersed in room-temperature distilled water. After 10 seconds, the final weight measurement was taken. The moisture on the film's surface was removed and weighed to calculate the amount of water absorbed. Repeat until the final weight is obtained with a constant sample. Eq. 1 was used to calculate the amount of moisture absorbed by each sample.

$$Water\ absorption\ (\%) = \frac{Final\ Weight\ (g) - Initial\ Weight\ (g)}{Initial\ Weight\ (g)} x\ 100\%$$

2.3.3. Biodegradation and characterization.

The biodegradation test examined the biodegradable, edible film properties, which were tested by burying the film in soil. In a container, the soil is prepared. All bioplastic films were cut into 2 cm x 2 cm squares and buried at a depth of 10 cm in a container filled with garden soil. The container was left inside the laboratory, and the soil was watered regularly to keep it moist. After a week, the film was taken out of the container, and the final weight was measured and written down. The functional groups present in the bioplastics produced were identified using FTIR analysis. The spectra were obtained in the 4000 cm^1 to 400 cm^1 range using an IR Prestige-21.

3. Results and Discussion

3.1. Tensile properties.

Tensile strength is a material's tensile strength, which is calculated by dividing the maximum force that the material can bear by the initial cross-sectional area of the material. The tensile strength at break is the maximum tensile strength that can be achieved before the plastic breaks or tears. The goal of tensile strength testing is to determine the tensile strength of bioplastics. Fig. 1 presents the effects of mangrove powder addition and concentrations of glycerol as plasticizing agents on bioplastics' tensile strength.

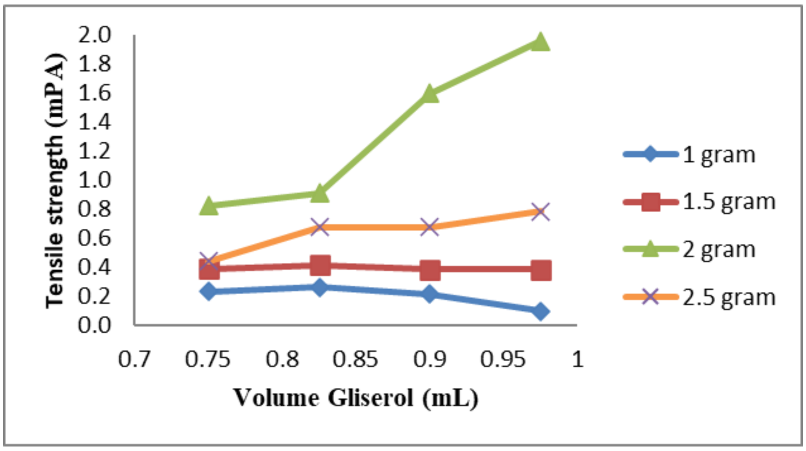

Figure 1. The effect of mangrove powder weight and glycerol on Tensile Strength (Mpa) of bioplastics.

From Figure 1, it can be seen that the bioplastic which has the highest tensile strength is the treatment using 2 grams of mangrove flour and the addition of 0.95 ml of glycerol, which is 1.958 Mpa, and the lowest is the use of 1 gram of mangrove fruit with the addition of 0.975 ml of glycerol, which is 0.101 Mpa. This is because the more glycerol is added, the more the tensile strength of the bioplastic will decrease. This decrease is related to the empty space that occurs because the bonds between polysaccharides are broken by glycerol. This causes the intermolecular bonds in the plastic to weaken and will result in plastic with lower tensile strength. In addition, plasticizers aided in the formation of hydrogen bonds between starch and plasticizer molecules. As a result, the tensile strength of the mangrove powder based on this study decreased due to the disruption and weakening of hydrogen bonds between the starch polymer chain [31]. The type of reinforcement and starch used can also increase the tensile strength of bioplastics.

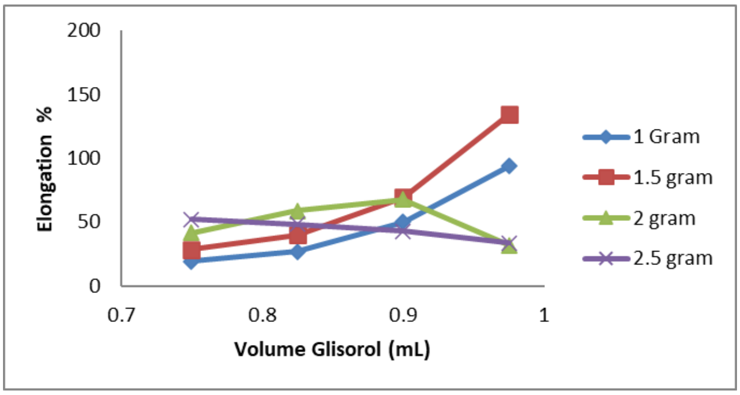

Figure 2. The effect of mangrove powder and glycerol on the percent elongation of bioplastics.

Because chitosan and starch have the function of increasing the mechanical properties of bioplastics, they can affect the tensile strength of bioplastics in this study. Moreover, adding chitosan resulted in significant antioxidant and antimicrobial activity, indicating [32]. This bioplastic made from mangrove fruit is possible and meets all the rules for making plastic bags for businesses.

On the other hand, elongation is a test performed in conjunction with a tensile strength test. The results of this test are used to calculate a percentage of the plasticity properties possessed by bioplastics. Research shows that a plastic film's extendibility from its initial length to its maximum breaking point is defined as elongation at break [33]. As shown in Figure 2, the highest percent elongation was found using 1.5 grams of mangrove flour and 0.75 ml of glycerol, which was 134.3%. Besides that, the lowest elongation percentage was found in using 1 gram of mangrove flour and 0.9 ml of glycerol, namely by 19.3%. It can be seen that the largest average elongation value is found in the addition of 0.75 ml of glycerol, and the lowest average is found in the addition of 0.975 ml of glycerol. The addition of glycerol aims to increase the plasticity of the bioplastic. Still, if the added glycerol is more than required, it can cause a decrease in the mechanical value of the bioplastic. This happens due to hydrogen bonds forming between the plasticizer and the starch molecules. As a result, the reconstruction of the molecular starch chains reduces crystallinity and thus increases the plastic film's flexibility.

3.2. Water absorption.

Water absorption is carried out to determine the ability to absorb water in the resulting bioplastic. The relationship between the effect of mangrove flour and the addition of glycerol to water content can be seen in Figure 3. This testing is essential for determining the stability of plastic film under humid and moist conditions. For this study, all bioplastic films were put into water that was distilled. This was to see how different concentrations of glycerol and mangrove powder changed the hydrophilic nature of starch plastic films. Figure 2. shows that the highest water absorption rate is found in the use of 2.5 grams of mangrove flour and the addition of 0.975 ml of glycerol, which is 49.94%, and the lowest water absorption is found in 1 gram of mangrove flour and the addition of 0.75 ml of glycerol, which is 13.82%. From the graphic data of the test results above, it can be seen that 2 grams of mangrove flour have lower water absorption than 1.5 grams of mangrove flour. This could be because, at the time of testing, the sample was not completely dried or left for a long time in an open environment; the bioplastic would absorb the surrounding water content.

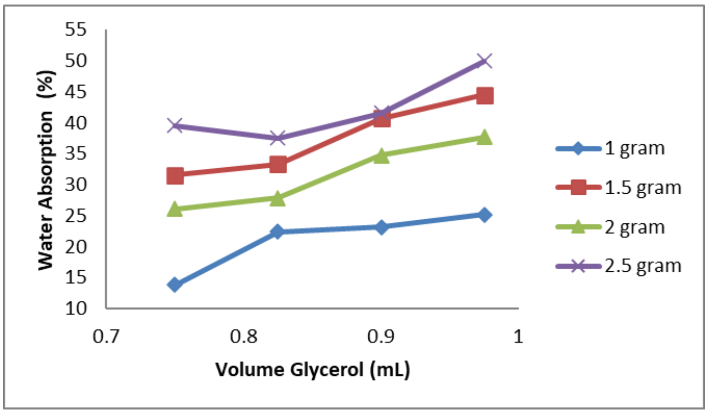

Figure 3. The effect of mangrove powder and the addition of glycerol on the percent water absorption.

Which should be the more mangrove flour and glycerol added, the higher the water content absorbed by the bioplastic. This is because glycerol is hydrophilic, absorbing water and binding water to bioplastics [34]. Bioplastics that are better at absorbing water aren't as good as bioplastics that are better at absorbing water because they can shorten the shelf life of bioplastics.

3.3. Soil burial test for biodegradability.

In order to see if microbial decomposers and soil moisture can break down the bioplastics and if they can be broken down by other things in the soil, this test will look at how resistant the bioplastics are to these things. The biodegradation of bioplastics is influenced by their physical and chemical structures, such as polymer chains, functional groups, and crystallinity, as well as the natural environment in which they are used (i.e., moisture, oxygen, temperature, and pH). Biodegradation is an enzymatic reaction catalyzed by microorganisms such as actinobacteria (Amycolatopsis, Streptomyces), bacteria (Paenibacillus, Pseudomonas, Bacillus, Burkholderia), and fungi in various ecosystems [35-39]. As a result, in Figure 4, burying bioplastic in soil media was used to conduct a bioplastic degradation test made from mangrove powder. The degradation took place in the soil for 7 days. The highest percentage of biodegradation was achieved with 2.5 grams of mangrove flour and 0.9 ml of glycerol, and the lowest percentage of biodegradation was achieved with 1 gram of mangrove flour and 0.975 ml of glycerol.

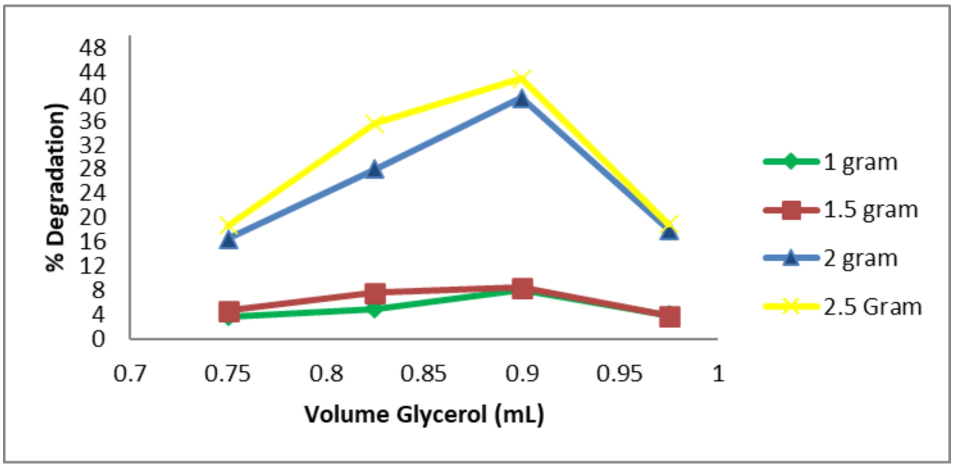

Figure 4. The effect of mangrove powder and glycerol on biodegradation of bioplastics.

According to these results, the best optimum percentage value was obtained by using 0.9 mL of glycerol in almost all variations in the composition of mangrove flour. Observations revealed clear degradation, specifically damage caused by fungi and decomposing bacteria. This is due to the presence of glycerol, which accelerates the degradation process and reduces the sample weight. Glycerol has hydrophilic properties that allow it to dissolve easily in water. Glycerol is taken up by the water in the soil. It has an impact on degradation.

3.4. Fourier transform infrared (FTIR) spectroscopy.

An Infrared (FT-IR) spectrophotometer was used to test the bioplastic functional groups. This analysis aims to determine the changes in the functional groups of a material or matrix that have been produced. This analysis is based on a wavelength analysis of a sample's characteristic peaks. Because each functional group has specific characteristic peaks, the wavelength of these peaks indicates the presence of certain functional groups in the sample. The FTIR spectra of this bioplastic were compared to different concentrations of mangrove powder and glycerol. Figure 4 depicts the FTIR spectra of the best bioplastic derived from mangrove powder and glycerol.

The spectrum shows that there is absorption at 3294.42 cm-1, evidenced by the presence of an N-H group. According to the literature, the absorption width observed in the 3300–3500 region is an N-H group absorption. While the absorption of the C-H group occurs in the area 3000–2850, the absorption of the O-H group occurs in the area 3600-2000, and the absorption of the C-O group occurs in the area 1690–1760. These findings point to the functional groups that comprise cellulose. Previous research on the synthesis of bioplastics from chitosan and starch from kapok banana peels with the addition of addictive substances yielded similar results, namely O-H, C-H, and N-H absorption in the formed bioplastics. The results of the functional group identification show that the overall functional groups that appear are the same as the basic ingredients used, cellulose (starch), which contains O-H, C-O, and C-H groups. Chitosan (functional groups O-H, C=O, N-H, and C-H) and glycerol (functional groups O-H, C-O). This means that no new functional groups have been found, which means that the resulting bioplastic has the same properties as its constituent parts and was made through a physical blending process.

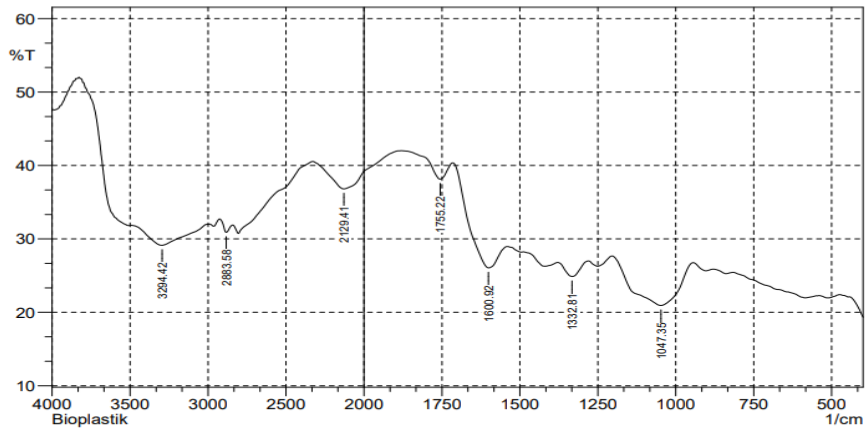

Figure 4. Graph of FTIR analysis of bioplastics on the best results of samples.

4. Conclusions

In this study, bioplastics based on mangrove starch powder and glycerol were successfully produced. The best bioplastics were discovered when 2 grams of mangrove flour and 0.9 ml of glycerol were combined, resulting in a tensile strength value of 1.6 Mpa, an elongation value of 67.9 percent, a water absorption capacity of 34.75 percent, a degradation percentage value of 39.8 percent, and an absorption group. NH (3294.5 cm^{-1}), CH (2883.5 cm^{-1}), OH (2129.41 cm^{-1}), and C=O are the functions (1755.22 cm^{-1}). The addition of glycerol increases the elongation value of bioplastics by up to 134% while decreasing the tensile strength by up to 0.386 mPA. The more mangrove flour and glycerol were used, the more water the bioplastics could hold. The peak degradation percentage for each variation of mangrove flour as a whole was reached with 0.9 ml of glycerol.

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Conflicts of Interest

The authors declare no conflict of interest.

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