EFFECT OF ADDITION OF CHITOSAN AND GLYCEROL ON BIOPLASTICS ORIGINATED FROM MELINJO SEEDS (Gnetum gnemon)

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Abstract

One solution to the plastic waste problem is to produce environmentally friendly plastic made from melinjo starch (Gnetum gnemon). The objective of this research is to determine whether bioplastic made from melinjo seeds (Gnetum gnemon) can be used as a starch-based material for producing eco-friendly plastic and to assess the impact of adding chitosan and glycerol to bioplastic derived from melinjo seeds (Gnetum gnemon). The research method employed is a qualitative and quantitative descriptive analysis of the resulting bioplastic, which consists of seven stages: starch production, bioplastic production, solubility test, swelling test, water resistance test, moisture content test, and biodegradability test. The results show that the composition with 4 grams of melinjo seed starch without the addition of chitosan and glycerol has low solubility (0%) and swelling (2.63%), high water resistance (97.37%), low moisture content (1.97%), and optimal biodegradation rate (97%). The addition of chitosan and glycerol affects the properties of bioplastic derived from melinjo seeds. This addition can increase solubility (2%), reduce the swelling percentage (1.47%), increase moisture content (3.11%), enhance water resistance (98.53%), and reduce the biodegradability percentage of bioplastic (97%).

Keywords: Bioplastic, Biodegradable, Glycerol, Chitosan, Starch

INTRODUCTION

Plastic bag waste in Indonesia is a serious environmental concern (Khumas et al., 2023). Research by Sustainable Waste Indonesia (SWI) shows that Indonesia produces more than 45.3 million tons of waste annually, with 15.6 million tons unmanaged and polluting the environment. Of this amount, 1.3 million tons is plastic waste (KLHK, 2018). Data from the National Waste Management Information System (SIPSN) of the Ministry of Environment and Forestry (KLHK) in 2022, based on input from 2021 regencies/cities across Indonesia, indicates that the national waste accumulation reached 21.1 million tons. Of this total national waste production, 65.71% (13.9 million tons) is managed, while the remaining 34.29% (7.2 million tons) is not properly managed (Kemenko PMK, 2023). According to data from the Ministry of Environment and Forestry (KLHK), the projected accumulation of plastic waste in Indonesia continues to increase from 9.2 million tons in 2017 to 9.9 million tons in 2025, equivalent to 13.98% of the total national waste volume. This increase is in line with the increase in national general waste generation. Indonesia is often cited as one of the world's largest producers of plastic waste, with ineffective waste management. The United Nations Environment Programme (UNEP) predicts that the amount of plastic waste entering marine ecosystems will nearly triple by 2040 if significant preventative measures are not taken (databoks, 2024).

Plastic produces carbon emissions when burned, polluting the environment. Carelessly discarded plastic waste can contaminate waterways and even accumulate in river floodgates, causing flooding. Furthermore, plastic waste disrupts fishing activities because its catch is filled with plastic waste, reducing catches, and fishing gear often suffers damage, such as torn nets and plastic waste getting caught in boat propellers. As a result, fishermen have to increase their fishing efforts, such as looking for better fishing locations or changing their routes (Sagita & Pratama, 2022).

One solution to the plastic waste problem is the production of environmentally friendly plastics called bioplastics (Maesaroh et al., 2021). Bioplastics are made from renewable biomass, such as microbiota, corn starch, fats, vegetable oils, and starch. Depending on the manufacturing process, plastics can degrade in both aerobic and anaerobic environments. In addition to biopolymers, cellulose, starch, and various other materials can be used as raw materials for bioplastics (Melani et al., 2017).

The technology for making biodegradable starch-based plastics has been developed in Indonesia for some time. Among the raw materials being studied for biodegradable plastic production are tapioca starch mixed with chitosan and glycerin (Setiarto, 2020), sago starch mixed with glycerol plasticizer, and corn starch (Purnavita & Dewi, 2021). However, commercially, the bioplastic production industry is still limited due to low domestic demand. Various studies have been conducted in Indonesia and other countries to explore the potential of biodegradable plastic raw materials (Yanuar, 2020).

Gnetum gnemon is a perennial plant that can grow for over 100 years, and each plant can produce 80-100 kg of melinjo (Farhan, 2024). The starch content of melinjo (Gnetum gnemon) seeds is 58.61%. The starch content in bioplastics can affect their strength and elasticity (Afif & Mursiti, 2018). The use of seeds containing starch has potential in bioplastic production (Ramadhan & Nugraha, 2021). One of the main requirements for bioplastic raw materials is a 50% starch content. Gnetum gnemon has a starch content of 58.61% (Anggraini et al., 2019).

Starch has significant potential as a raw material for bioplastic production due to its renewable nature and biodegradability (Suryani et al., 2022). Starch is used as the primary ingredient in bioplastic production due to its environmentally friendly, readily degradable, abundant, and affordable nature (Melani et al., 2022). The starch content in bioplastics varies. Starch from cassava peels, fruit seeds, and banana peels has varying starch content, starting at 50% (Melani et al., 2019).

Starch-based bioplastics have potential for development, but they suffer from drawbacks such as poor mechanical properties and less water resistance compared to traditional plastics. However, these issues can be addressed by adding fillers, plasticizers, and other materials. Plasticizers increase the flexibility and strength of bioplastics by reducing interactions between polymer chains. Glycerin is one such material that can be used as a plasticizer.

The addition of glycerol during bioplastic production can improve the mechanical properties and water resistance of bioplastics. Chitosan is a polymer made from the deacetylation of chitin, a polymer compound found in the shells of shrimp, shellfish, yeast, insects, and fungi (Yuniarti & Hatina, 2021). Research on chitosan bioplastics from sugar palm (Arenga pinnata) has shown that chitosan improves the mechanical properties and water resistance of bioplastics (Suwardi & Hidayati, 2020). Furthermore, research.

Based on the above background, research is needed to investigate the effect of adding chitosan and glycerol to bioplastics derived from melinjo seeds (*Gnetum anemon*).

METHODS

Making Melinjo Seed Starch (Gnetum gnemon)

Gnetum gnemon (Gnetum gnemon) seeds were weighed as much as 200 grams (every 100 grams of melinjo seeds produce 48 grams of starch), washed and cut into several pieces. Then, the melinjo seeds (Gnetum gnemon) were blended with distilled water in a 1:1 ratio (200 grams of melinjo seeds added to 100 ml of distilled water) until they had a porridge-like texture. After blending, they were filtered using a filter cloth and

left to settle for 24 hours. The melinjo seed dregs were washed 3 times using distilled water. The filtered dregs were left to settle for 24 hours. The filtered results were dried using an oven at 50 °C for 6 hours. After drying, the starch was ground using a mortar and filtered using a 100-mesh sieve (Pratiwi & Widodo, 2021). The composition of the bioplastic production is in the table (Table 1).

Table 1 Composition of starch, chitosan, and glycerol in the treatment

No	Treatment	Composition		
		Pati (g)	Kitosan (g)	Gliserol (%)
1	P1	4	0	0
2	P2	4	1	15
3	P3	4	1,5	35
4	P4	4	2	55

Making Melinjo (Gnetum gnemon) Seed Bioplastic

In this study, bioplastic was made using a blending method, a method of mixing two or more materials into one (Alfarisi et al., 2021). The bioplastic film was synthesized by dissolving chitosan (1 gram, 1.5 grams, and 2 grams) in 100 milliliters of 3% acetic acid (Nafianto, 2019). This process was carried out for 30 minutes using a magnetic mixer until homogeneous. Afterward, 4 grams of melinjo (Gnetum gnemon) seed starch was dissolved in distilled water. Glycerol, according to the treatment (15%, 35%, and 55%), was added to the starch by heating on a hot plate for 12 minutes. The two solutions were mixed by heating the hot plate at 70°C for 1 hour. The bioplastic mixture was then poured into a plastic mold. The bioplastic was removed from the mold and ready for testing after being left at room temperature for six days (Ermawati & Haryanto, 2020).

Solubility Testing

The solubility testing followed the Sharma and Singh method, which involved cutting the bioplastic into 20 x 20 mm pieces, each sample being treated once. The samples were then oven-dried at 50°C for 30 minutes. The samples were then weighed to determine their initial weight (W0). Subsequently, the samples were immersed in 5 ml of distilled water for 1 minute at room temperature. The soaked samples were then oven-dried at 100°C for 45 minutes. The samples were then weighed to determine their final weight (W1). The solubility of the bioplastics was calculated using the formula (Wijayani et al., 2021).

$$Kelarutan (\%) = \frac{(W0 - W1)}{W0} \mathbf{x} \mathbf{100}$$

Swelling Test

Using the gravimetric method. The water resistance of plastic films is determined by the swelling test, which is the percentage of film swelling due to the presence of water. The lower the water absorption value, the better the plastic properties, while the higher the water absorption, the more easily damaged the plastic properties (Budiman et al., 2018). The swelling percentage test is carried out using the method used, namely by cutting the sample with a size of 20x20 mm. Then, the sample is weighed to obtain the initial weight value (W0). After that, the sample is immersed in 15 ml of distilled water for 1 minute. The sample is removed and wiped with a tissue. After that, the sample is weighed to obtain the final weight value (W). The swelling test is calculated using the following formula (Hasri, et al. 2021).

Swelling (%) =
$$\frac{W1 - W0}{W0}$$
 x 100%

Water Resistance Test

The water resistance test uses a gravimetric method, where the weight of the bioplastic is measured before and after treatment (Purnavita & Dewi, 2021). The water resistance value is calculated using the formula (Hasri et al., 2021).

Water Resistance = 100 - nilai % swelling swelling

Water content test

Bioplastic moisture content testing is carried out by oven drying, using a gravimetric method where the weight of the bioplastic before and after treatment is weighed (Purnavita & Dewi, 2021). The test is carried out by drying the porcelain cup used in an oven at 105°C for 30 minutes, then removing the cup and placing it in a desiccator for 15 minutes and weighing its weight (A). After that, a 1 gram sample is weighed and placed in a porcelain cup (B) and then dried in an oven at 105°C for 3 hours. Then, it is placed in a desiccator for 15 minutes and weighed (C). This test is modified by weighing the sample. Weight loss is calculated as a percentage of water content and is calculated using the formula (Yuniastuti & Muryeti, 2021).

Kadar air (%) =
$$(B-C)(B-A)x$$
 100

Degradation Test (Biodegradability)

The method used to observe biodegradability characteristics is the soil burial test (Ridwan, 2018). This test indicates that the plastic is fully degraded based on the percentage weight loss (% weight loss) up to 100% (Marlina & Nurhalizah, 2021). Bioplastic samples were tested to determine the level of plastic degradation. The decomposition time was calculated by observing them periodically every four days. Visual observations were made every four days for 16 days. If the bioplastic can be decomposed by microorganisms or degrades well, it is considered environmentally friendly. In this study, bioplastic samples measuring 2 x 2 cm were used to assess biodegradability (Ermawati & Haryanto, 2020).

$$Biodegradabillity = \frac{Berat\ awal-Berat\ akhir}{Berat\ awal}x\ 100\%$$

RESULTS AND DISCUSSION

Hasil Uji Kelarutan Bioplastik Biji Melinjo (Gnetum gnemon)

The solubility test results for treatment P2 showed the highest solubility at 33%, followed by P3 and P4 at 17% each, and P1 at 0% (P1). The lowest solubility (P1) is the best bioplastic result because it does not dissolve easily in water:



Figure 1. Graph of Bioplastic Solubility Test Results

This research aligns with previous research by Umi Ermawati and H Haryanto (2020), which showed that the addition of chitosan can improve the mechanical properties and biodegradability of bioplastics. This study found that the addition of chitosan at certain concentrations strengthens the bioplastic, making it more resistant to degradation, which indirectly reduces solubility. These results are similar to those found in treatment P1, where low solubility indicates the bioplastic's resistance to dissolution in water.

The addition of glycerol can increase the water solubility of bioplastics. This research aligns with research by Aripin et al. (2017), which showed that the addition of glycerol can reduce intermolecular forces in bioplastics, resulting in increased solubility. Furthermore, research by Budiman et al. (2018) also stated that the higher the glycerol concentration, the higher the water solubility percentage. The water-soluble properties of biodegradable plastic films are influenced by the presence of -OH groups derived from glycerol.

Starch is known for its hydrophilic properties, meaning its tendency to absorb water (Syahrum et al., 2017). Molecularly, starch consists of two main components: amylose and amylopectin (Sari et al., 2020). Amylose is a linear, water-soluble glucose polymer, while amylopectin is a less soluble, branched polymer (Abdillah, 2022). The combination of these two components gives starch properties that influence the behavior of bioplastics. Starch has a significant drawback when used as a bioplastic material: its slow dissolution in water at room temperature (Setiarto, 2020).

This is consistent with research by Nafianto (2019) on water absorption in bioplastics from kepok banana stem waste, which showed that starch content significantly influences the water absorption capacity of the resulting bioplastic. Increasing starch content generally reduces the water absorption capacity of bioplastics in room-temperature water tests.

Swelling Test Results of Melinjo Seed Bioplastic (Gnetum gnemon)

The swelling test results for melinjo (Gnetum gnemon) seed bioplastics showed that treatment P1 had the highest swelling of 2.63%. Treatment P4 had the lowest swelling of 1.47%. Furthermore, treatment P2 had a swelling of 2.08%, and treatment P3 had a swelling of 72%. The results showed that the addition of chitosan and glycerol reduced the swelling percentage. Therefore, it can be concluded that a composition of 4 grams of starch, 2 grams of chitosan, and 55% glycerol (P4) was the best composition, achieving the lowest swelling value of 1.47%.

Furthermore, research by Indra Nafianto (2019) also showed that increasing the chitosan content reduced swelling in bioplastics made from kepok banana stems. This study confirms the finding that chitosan plays a role in reducing swelling. However, this study also found that increasing the glycerol volume actually increased swelling, in contrast to the swelling test results for melinjo seed bioplastics, which showed a decrease in swelling with the addition of glycerol.

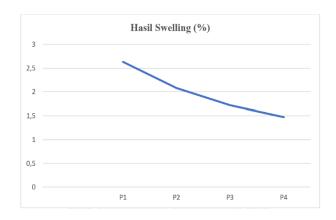


Figure 2 Swelling Test Results Graph

The results of the Melino (Gnetum gnemon) seed swelling test showed that the addition of chitosan can reduce the swelling percentage. These results are similar to the research of Yuniastuti & Muryeti (2021), which revealed that chitosan can reduce swelling in bioplastics. The hydrophobic nature of chitosan reduces the ability of bioplastics to absorb water, thus reducing swelling. Fajarwati (2020) stated that chitosan helps create a denser and less permeable structure to water, thereby reducing the expansion of the thickness of bioplastics when exposed to water. In addition,

Water Resistance of Melinjo Seed Bioplastic (Gnetum gnemon)

The water resistance test results for melinjo (Gnetum gnemon) seed bioplastics showed that treatment P4 had the highest water resistance at 98.53%. Treatment P1 had the lowest water resistance at 97.37%. Furthermore, treatment P2 had a water resistance of 97.92%, and treatment P3 had a water resistance of 98.28%. Therefore, it can be concluded that the composition of 4 grams of starch added to 2 grams of chitosan and 55% glycerol (P4) is the best composition because it achieved the highest water resistance value of 98.53%.

Previous research by Umi Ermawati and H. Haryanto (2020), which used jackfruit seeds as the main ingredient, showed similarities in the use of chitosan and glycerol as components that improve the mechanical properties of bioplastics. This study showed that the addition of chitosan and glycerol had a positive effect on the tensile strength and elongation of bioplastics, although it did not specifically measure water resistance.

Previous research by Indra Nafianto (2019) using kepok banana stem waste, chitosan from snail shell waste, and glycerol from used cooking oil also found that variations in the composition of chitosan and glycerol affected the characteristics of bioplastics, including tensile strength and elongation.

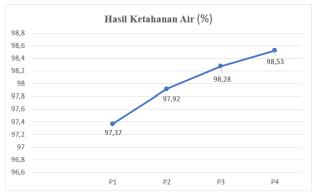
The results of this study revealed that the addition of chitosan and glycerol can increase water resistance. The addition of chitosan increases hydrogen interactions between chitosan, glycerol, and starch molecules, thereby reducing hydrogen interactions between water molecules, ultimately increasing water resistance.

Water Resistance Test Results of Melinjo Seed Bioplastic (Gnetum gnemon)

The results of the water content test for melinjo (Gnetum gnemon) seed bioplastic showed that treatment P4 had the highest water content, at 3.11%. Treatment P1 had the lowest water content, at 1.97%. Furthermore, treatment P2 had a water content of 2.89%, and treatment P3 had a water content of 3.02%. Increasing the glycerol concentration can increase the water content of the bioplastic. Compared to research by

Ermawati and Haryanto (2020), which examined the effect of chitosan and glycerol addition on the characteristics of bioplastic films made from jackfruit seed starch, water content was not the primary parameter discussed. That study focused more on the tensile strength, elasticity, and biodegradability of bioplastics with varying chitosan and glycerol concentrations.

Furthermore, Nafianto's (2019) research on the production of biodegradable plastic from kepok banana stem waste with glycerol and chitosan plasticizers did not specifically focus on water content, but included tensile strength and elongation tests, as well as the effect of chitosan and glycerol composition on bioplastic properties. The results of this study indicate that increasing the chitosan composition reduces water absorption and slows the bioplastic degradation process, while increasing the glycerol concentration accelerates bioplastic degradation. According to Polnaya et al., (2023), the addition of glycerol can increase the hydrophilicity of bioplastics by increasing the OH groups of glycerol, thereby increasing the amount of water bound. This statement is consistent with the research of Rifaldi & Bahruddin (2017) which shows that the addition of glycerol can increase the percentage of water content. Research by Novita & Rahmadhia (2021) also states that the addition of glycerol can increase the water content of bioplastics.



Gambar.3 Grafik Hasil Ketahan Air Bioplastik

The hydrophilicity of bioplastics increases with the addition of OH groups from glycerol, which increases the amount of water bound. This statement aligns with research by Rifaldi & Bahruddin (2017), which showed that adding glycerol can increase the water content. Research by Novita & Rahmadhia (2021) also indicated that adding glycerol can increase the water content of bioplastics.

Biodegradation Test Results of Melinjo Seed Bioplastic (Gnetum gnemon)

The results of the biodegradation of melinjo seed bioplastic (Gnetum gnemon) showed that treatment P1 showed the highest biodegradation value of 97% and P2, P3 and P4 showed the lowest Biodegradation value of 94%. The addition of glycerol and chitosan glycero can affect the biodegradation of bioplastics. The addition of chitosan in bioplastics can slow down the biodegradation of bioplastics due to the low level of damage and degradation will take longer. Thus, it can be concluded that the composition of 4 grams of starch without the addition of glycerol and chitosan (P1) is the best degradation result of 97%.

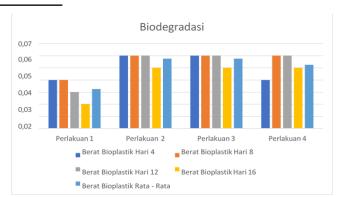


Figure .5 Average Graph of Bioplastic Biodegradation Observations

Previous research by Umi Ermawati and H. Haryanto (2019) showed that the addition of glycerol and chitosan affected the biodegradation rate of the resulting bioplastic, with chitosan slowing down biodegradation. Research by Indra Nafianto also explored the effect of chitosan and glycerol addition on the mechanical properties and biodegradation of bioplastics from banana seed starch, showing that glycerol increased elongation and chitosan affected tensile strength. Furthermore, previous research by Umi Ermawati and H. Haryanto (2020) tested varying amounts of chitosan and glycerol in the production of bioplastics from jackfruit seed starch and found that the addition of 1 gram of chitosan and 15% glycerol produced the best tensile strength, while the highest biodegradation occurred with different combinations of chitosan and glycerol.

This study demonstrates that the addition of glycerol and chitosan affects the biodegradation of melinjo (Gnetum gnemon) seed bioplastics. This is in accordance with research by Permata & Imam (2023) which stated that the best biodegradation was without chitosan. According to Hendrawati (2023), the addition of chitosan can slow the decomposition of bioplastics. This is due to the hydrophobic nature of chitosan and its insolubility in water. By adding chitosan to the bioplastic solution, the level of damage decreases and the degradation time becomes longer. This is related to the ability of biodegradable plastics to absorb water. The higher the content of a particular material, the faster the material degrades. According to Setiarto (2020), the degradation of biodegradable plastics is influenced by several factors, including microbes, soil moisture, soil type, and the properties of the biodegradable plastic itself.

CONCLUSION

Gnetum gnemon seeds are effectively used as a basic ingredient in the manufacture of environmentally friendly plastics, especially when combined with the right composition of chitosan and glycerol. The results showed that the best composition for water solubility testing was P1 (0%), swelling testing was P4 (1.47%), water resistance testing was P4 (98.53%), water content (3.11%) and biodegradation was P1 (97%). The addition of chitosan and glycerol affects the properties of bioplastics derived from gnetum gnemon seeds. This addition can increase solubility, decrease swelling, increase water content, increase water resistance and reduce biodegradation of bioplastics.

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