



# Biodegradable Plastic Synthesis Based on Kluwih Seeds (*Artocarpus camansi*) and Chitosan with the Addition of Glycerol

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**Abstract:** The natural resources of agricultural products have the potential to be raw material for making biodegradable plastic, one of which is starch from kluwih seeds (*Artocarpus camansi*). This study examines the potential of kluwih seed starch as a basic material for making biodegradable plastic. This research method is experimental, the raw material for making biodegradable plastic is the kluwih seed starch originating from Aceh Besar Regency, Aceh Province. Other additives used are chitosan as reinforcement and glycerol as plasticizers. The analysis is carried out, namely mechanical properties, morphology, thermal, functional groups, water absorption and degradation analysis. The results of the analysis of this study show that the kluwih seed starch functions as a basic material for making biodegradable plastic. As chitosan concentration increases, the resulting tensile strength increases with results obtained around 119.17-166.67 kgf/mm<sup>2</sup>, while the percentage of elongation is lower the results obtained are 6.82-11.15%. The morphological analysis shows the biodegradable plastic produced has a compact and homogeneous structure. The thermal analysis shows a good thermal stability to ranging from 180 to 370 °C. The results of the biodegradable plastic FTIR show wave numbers 1662.64–1049 cm<sup>-1</sup>. Water absorption analysis is obtained around 7.658-12.068%, a decrease in water absorption is influenced by the addition of chitosan concentration. Biodegradable plastic planted in the soil containing EM4 degraded for 3 days.

**Keywords:** Biodegradability; Chitosan; Glycerol; Kluwih; Starch

## Introduction

Packaging currently has an important role for products in terms of maintaining consumer safety. Packaging functions can protect products from oxygen, water vapor and contamination (Maliha et al., 2020). Packaging is currently widely used to package household appliances, electronics, food and beverages. Lack of conventional packaging is difficult to degrade so as to damage the environment (Cahyaningtyas et al., 2019). As an increase in the use of plastic, it causes increased buildup of waste so that it has a negative impact on the environment, both environmental

pollution and interferes with the lives of various organisms (Lan et al., 2021). Plastic packaging is difficult to degrade due to raw materials for making petroleum (Briassoulis & Mistriotis, 2018; Briassoulis et al., 2020; Lee & Liew, 2020), recycling plastic waste is less effective and very expensive (Triawan et al., 2020). Biodegradable plastic is an alternative in reducing the use of conventional plastic that can reduce environmental pollution. One of the uses of biodegradable plastic is suitable to be applied to food and non-food packaging (Tavassoli-Kafrani et al., 2015). Biodegradable plastic is made from organic sources such as starch, cellulose and chitin (Savadekar et al., 2015). The biopolymers are

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extracted from agricultural products such as starch, protein and cellulose (Santana et al., 2018).

Starch is a semicrystalline hydrophilic biopolymer consisting of a mixture of two glycosidic macromolecules namely amylose and amylopectin in the form of granules (Roy et al., 2019). As for the advantages of starch because it is available throughout the year (renewable), flexibility, and natural decomposition (biodegradable), starch consists of two glycosidic macromolecules, namely amylose and amylopectin (González et al., 2015). The use of starch as raw material for making biodegradable plastic has been carried out by previous researchers, including starch from corn (Lin et al., 2019), mango skin (Adilah et al., 2018), Purple Sweet Potato (Jiang et al., 2020), cassava tubers (Hidayat et al., 2020; Sirisha et al., 2020). Amylose levels in starch range from 15 to 30%, while amylopectin is 70 to 85%. The higher the amylose level can improve the biodegradable plastic mechanical properties produced (Cahyaningtyas et al., 2019). Starch sourced from kluwih seeds (*Artocarpus camansi*) has an amylose content of 30% (Hidayat et al., 2023). In this research, biodegradable plastic was made from starch sourced from kluwih seed extract with the aim of improving the mechanical properties of the biodegradable plastic produced.

## Method

### Material

Kluwih seed starch (*Artocarpus camansi*) obtained from the local market (Indrapuri Regency of Aceh Besar), chitosan, glycerol, 2% acetic acid and aquadest. While the tools used for the characterization of tensile strength and elongation use the Standard Test Method for Tensile Properties of Thin Plastic Settings ASTM D882-09 (ASTM, 2009). Functional group analysis using Fourier Transform Infrared (FTIR) Spectrometer (Shimadzu Prestige-21). Morphological analysis using microscope (SEM) electron scanning (Jeol JSM-6510 LA). While thermal analysis uses Shimadzu devices (TGA-503).

### The Process of Making Biodegradable Plastic

In the manufacture of biodegradable plastic in this study using the principle of thermodynamics, the solidification process of the liquid phase to solid. There are 2 (two) stages in the manufacture of biodegradable plastic, the one chitosan stage is dissolved into a 2% acetic acid solution using magnetic stirrer for 1 hour, then filtered using filter paper. The second stage of weighing the starch produced from kluwih seeds (*Artocarpus camansi*) and dissolved into 100 ml of distilled water, then added chitosan solution. The

mixture is stirred using a hot plate and magnetic stirrer at a speed of 60 rpm with a temperature of 60 °C for 2 minutes. Next is added glycerol as a plasticizer, then stirred for 10 minutes after it was cooled. The results obtained are poured into a glass mold with a size of 30 cm x 20 cm in a rectangular form and leveled. After being flat, the mold results are dried for 24 hours at room temperature. Then put into a desiccator, the biodegradable plastic is ready to be analyzed.

### Biodegradable Plastic Characteristics Analysis of Mechanical Properties

Tensile strength (kgf/mm<sup>2</sup>), percent of breaking extension (%) and modulus young (KGF/mm<sup>2</sup>) are measured using texture analysis (Ta -XT2I, Stable Micro Systems, Surrey, UK) which operates according to ASTM D882-09 (ASTM, 2009). Ten strips of each film are used.

### Microscope Electron Scanning Analysis

Biodegradable plastic was previously conditioned in a desiccator (0%RH) containing P<sub>2</sub>O<sub>5</sub> (25°C) for 3 weeks. Film pieces are broken down in liquid nitrogen. Before observations, the sample is mounted on the metal grid using a two-sided adhesive tape and coated with gold in a vacuum condition. Observation of micro-sectional structures is done using an electron microscope scanning (SEM Jeol 6510 LA-Japan) at 5 kV.

### TGA Analysis

Thermogravimetric Analysis (TGA) Biodegradable plastic uses Shimadzu devices (TGA-503, Brazil). The sample is heated, using a nitrogen atmosphere, from room temperature up to 800 °C at a rate of 20 °C min<sup>-1</sup>.

### FTIR analysis

Analysis of Fourier Transform Infrared Spectroscopy (FTIR) Plastic Biodegradable using Shimadzu Prestige-21. The sample is dried in a desiccator containing silica gel before analysis. Sample pieces of 2 cm in diameter are clamped between two KBr disks. FTIR spectrum recorded from wave numbers 400-4000 cm<sup>-1</sup>.

### Water absorption analysis

Analysis of water absorption is carried out to determine the amount of water that can be absorbed by biodegradable plastic. Biodegradable plastic samples are cut with size 2 cm x 2 cm. Then weighed as the initial mass (W<sub>0</sub>). Then put in a glass containing distilled water for 10 seconds then removed from the glass filled with distilled water and placed on filter paper. Re-weigh the biodegradable plastic mass (W).

The value of biodegradable plastic resistance to water is calculated using the equation:

$$\text{Water absorption}(\%) = \frac{W - W_0}{W_0} 100\% \quad (1)$$

Description:

W = Biodegradable plastic mass after immersion

W<sub>0</sub> = Early mass of biodegradable plastic

*Biodegradable Plastic Analysis*

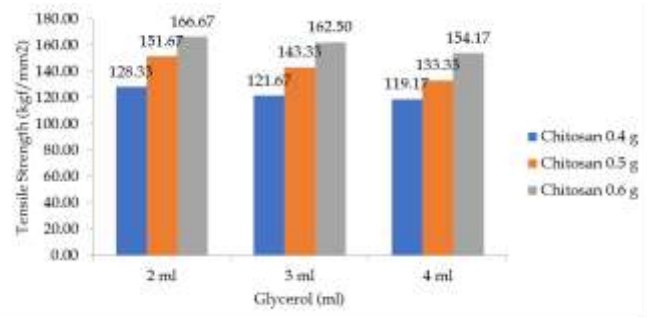
Analysis conducted includes mechanical properties including tensile strength and elongation, thermal, functional groups, morphology, water absorption and biodegradability analysis that aims to determine the length of time needed to break down biodegradable plastic naturally.

**Result and Discussion**

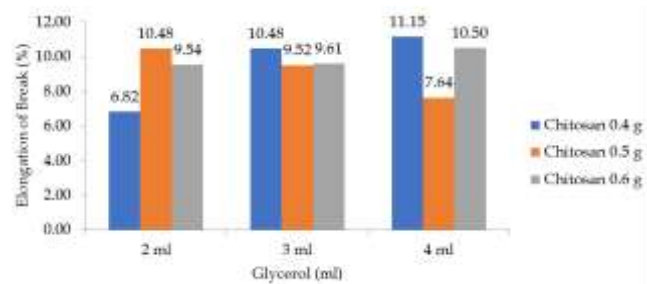
*Mechanical Analysis*

Testing the mechanical properties of biodegradable plastic aims to determine the amount of force achieved by the maximum pull on a unit area of biodegradable plastic to stretch or elongate. In Figure 1 it can be seen that the tensile strength of biodegradable plastic is highest in the treatment of 0.6 g Chitosan with the addition of 2 mL glycerol with a tensile strength value of 166.67 kgf/mm<sup>2</sup>. The tensile strength increases as the chitosan concentration increases. Meanwhile, the lowest tensile strength value was biodegradable plastic treated with 0.4 g Chitosan with the addition of 4 mL glycerol with a tensile strength value of 119.17 kgf/mm<sup>2</sup>. According to Zheng et al. (2019) the decrease in the mechanical properties of biodegradable plastics is caused by intra-molecular hydrogen bonds. In addition, the composition of chitosan, the presence of plasticizers and emulsification have an impact on the mechanical properties of biodegradable plastics, causing variations in mechanical properties (Peng & Li, 2014).

Elongation is the elongation of break of biodegradable plastic. Figure 2 shows that the elongation value of biodegradable plastic ranges from 6.82 to 11.15%. The highest elongation percentage value at a chitosan concentration of 0.4 g with a glycerol concentration of 4 mL was 11.15 %. In Figure 2, it can be seen that increasing the concentration of glycerol can increase the elasticity of the biodegradable plastic produced. This is because the hydroxyl groups contained in glycerol can form intermolecular hydrogen bonds with the hydrophilic groups in biodegradable plastic based on kluwih seed starch with the addition of chitosan. The higher levels of added chitosan inhibit the movement of polymer chains and reduce their flexibility (Jiang et al., 2020). Adilah et al. (2018) added that the percent elongation of biodegradable plastic is influenced by the addition of large amounts of chitosan and starch.



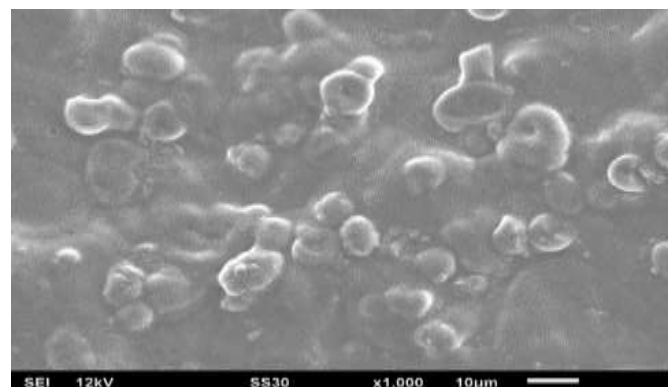
**Figure 1.** Tensile strength of biodegradable plastic



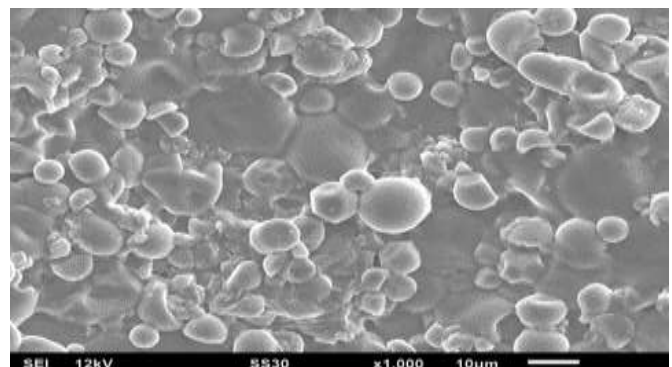
**Figure 2.** Elongation value of biodegradable plastic

*Scanning Electron Microscope (SEM) Analysis*

Observation of the morphology of biodegradable plastic was seen using an electron microscope (Scanning Electron Microscope, SEM). This analysis aims to explain the morphology of the biodegradable plastic produced.



**Figure 3.** SEM with a chitosan concentration of 0.4 g



**Figure 4.** SEM with a chitosan concentration of 0.5 g



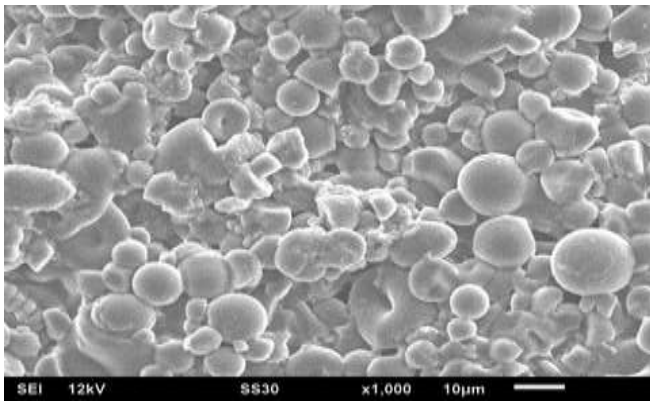


Figure 5. SEM with a chitosan concentration of 0.6 g

SEM results of biodegradable plastic with a composition of kluwih seed starch and chitosan with the addition of glycerol as a plasticizer with a magnification of 1000×, in Figure 3 with the addition of 0.4g chitosan it can be seen that the surface of the biodegradable plastic is not homogeneous, whereas in Figure 4 with the addition chitosan concentration of 0.5g shows that the kluwih seed starch granules in biodegradable plastic are not solid and are still hollow, in Figure 5 with the addition of 0.6g chitosan it can be seen that the kluwih seed starch granules tend to be denser. According to Romainor et al. (2014), as more chitosan is added, the granules become denser, this is due to the cross-linking network between chitosan. With the addition of chitosan which functions as a reinforcement, it is difficult for gas to penetrate and can increase the tensile strength value (Hidayat et al., 2020).

*Thermal Analysis*

Thermalgravimetric test (TGA) was carried out on optimal samples to determine the thermal stability of the resulting biodegradable plastic.

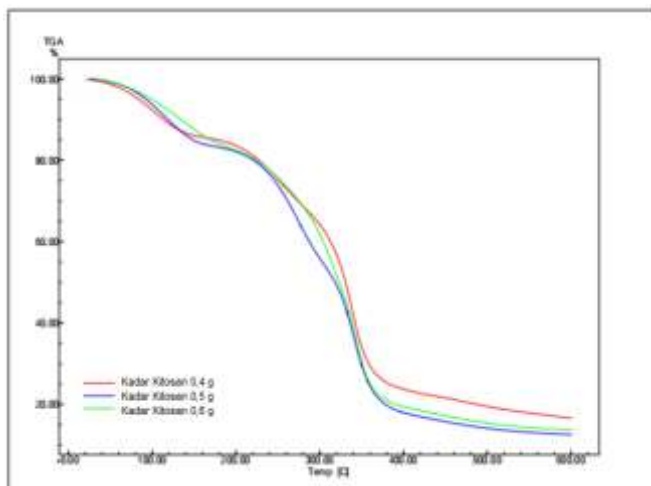


Figure 6. TGA analysis results for samples representing combined biodegradable plastic curves

Based on the figure, in the first stage of the curve for biodegradable plastic with chitosan content of 0.4 g and 0.5 g, a small decrease in primary mass was observed at temperatures from 140 to 370 °C, while in the second stage of the TGA curve it can be seen with the increase in chitosan concentration. 0.6 g of mass decrease occurs at temperatures from 180 to 370 °C, this is due to evaporation of water vapor and volatile substances. According to Soltani et al. (2018), increasing temperature results in evaporation and decomposition of primary moisture starting from 160 °C and continuing up to 385 °C. In addition, biodegradable plastics with the addition of glycerol can reduce the Tg value, possibly due to the ability of glycerol to penetrate the polymer chains and therefore weaken the interaction between polysaccharides and chitosan (Hosseini et al., 2013). Meanwhile, the higher the molecular weight and chain length, the higher the temperature required (Jimenez et al., 2013). Based on the TGA curves of Figure 6, both samples show good thermal stability up to 100 °C, although the kluwih seed starch-based sample shows a faster decomposition rate at higher temperatures.

*FTIR Analysis*

Fourier transform infrared spectroscopy (FTIR) analysis is one of the techniques used to identify chemical components, in identifying functional groups of biodegradable plastics in the range of 500-4000 cm<sup>-1</sup>. FTIR spectroscopy of biodegradable plastic can be seen in Figure 7.

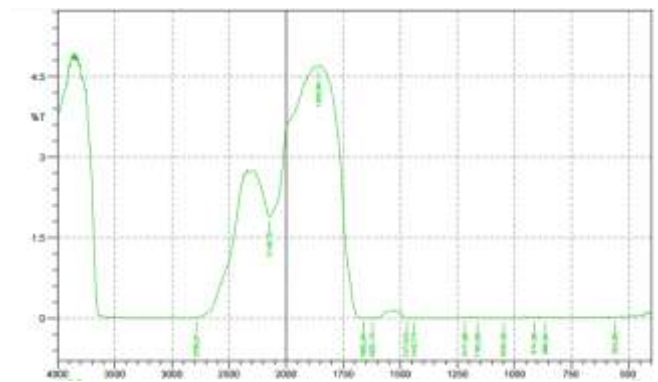


Figure 7. Functional groups of biodegradable plastic

**Table 1.** Biodegradable Plastic Functional Groups

| Wave number (cm <sup>-1</sup> ) | Functional groups        |
|---------------------------------|--------------------------|
| 1662.64                         | Vibration C=O            |
| 1217                            | Vibration C-O            |
| 1163                            | Vibration C-N (amine)    |
| 1049                            | Vibration C-H (aromatic) |

Figure 7 and Table 4 show that the wavelengths in FTIR analysis can provide information about the chemical composition of biodegradable plastics. Some wavelengths are closely related to the components or

chemical groups present in biodegradable plastic, such as the wave number  $1662.64\text{ cm}^{-1}$  which indicates the vibration of the C=O carbonyl bond. In biodegradable plastics, this can be related to the presence of polymers containing carbonyl groups, such as polymers made from starch. At  $1471\text{ cm}^{-1}$  this wavelength is often related to the vibrational strain of C-H bonds in aromatic compounds. In compounds containing aromatic rings, CH vibrations often occur around this wavelength  $1442\text{ cm}^{-1}$ . This range can also be related to the vibrational strain of the CH bond, especially to the CH<sub>2</sub> vibration (methylene group) or also to the asymmetric COO- (carboxylate group) vibration.

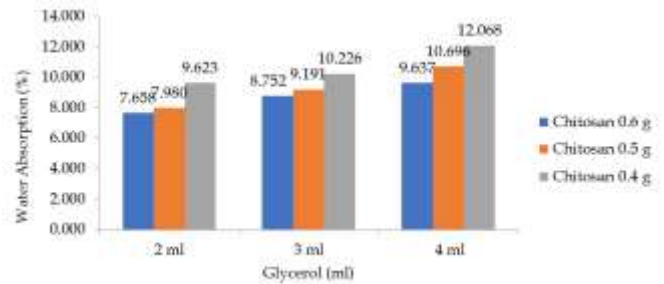
At  $1217\text{ cm}^{-1}$  is the vibration of the C-O bond in alcohol, ether or ester compounds. This could be related to the presence of ester groups which are commonly found in several types of biodegradable plastic, such as polyester which is made from polylic acid. The wave number  $1163\text{ cm}^{-1}$  indicates the vibration of the C-N bond in the amine compound. Although not often used in biodegradable plastics, some types of biodegradable plastics can use materials that contain amine groups in their composition. Next,  $1049\text{ cm}^{-1}$  is C-O stretching in aromatic compounds. The C-O functional group of C-O-C with a sharp absorption band and strong intensity is in the fingerprint area with a wavelength of  $1049\text{ cm}^{-1}$  and at a wavelength of  $914$  which is the C-O fringer print area in alcohol compounds (Bartolucci et al., 2023; Haghghi et al., 2019; Yupa et al., 2021).

*Water Absorption Analysis*

The water absorption capacity of biodegradable plastic is determined based on the Swelling Index which can be shown in Figure 8. Based on the results of this research, it can be seen that the greater the concentration of chitosan, the more water absorption capacity of biodegradable plastic tends to decrease. In Figure 8, you can see that the chitosan concentration of 0.6g with the addition of 2 mL of glycerol produces the lowest value of 7.658%. This is due to the hydrophobic nature of chitosan, so it can reduce the swelling of the resulting biodegradable plastic (Shahbazi, 2017).

Meanwhile, adding a concentration of 4 mL of glycerol and 0.4 g of chitosan had the highest water absorption value for biodegradable plastic at 12.068%. The higher the addition of glycerol, the higher the water absorption percentage. This is because glycerol also has more hydroxyl groups which have polar properties. Water absorption capacity can also be related to the chemical structure of materials that have functional groups (OH) that can adsorb water (Hidayat et al., 2020). The intrinsic swelling properties of biodegradable plastics with the addition of chitosan and glycerol are related to the presence of hydrophilic groups such as

carboxylic groups in their structure, which can easily interact with water (Kakaei & Shahbazi, 2016). According to Shahbazi (2017) and Wu et al. (2015) biodegradable plastics can be influenced by water diffusion, polymer linkages, the amount of hydrogen and ionic bonds as well as the nature of intermolecular chain interactions.



**Figure 8.** Swelling index

*Biodegradable Analysis*

Biodegradation analysis of biodegradable plastic was carried out by observing the decomposition process in the soil which was characterized by a reduction in the mass of biodegradable plastic when planted in compost soil. In this research, it was carried out by burying them in soil media containing EM4 bacteria (Effective Microorganisms) and samples were observed every day. Biodegradable plastic samples were tested by burial in soil containing EM4 bacteria, on the third day all samples had completely decomposed. This can be shown by the damage to the surface of biodegradable plastic.



**Figure 9.** Biodegradable plastic is burying in soil media containing EM4 bacteria (Effective Microorganisms)

## Conclusion

Kluwih seed starch can be used as a basic material for making biodegradable plastic, even though the tensile strength produced is not yet optimal, but the SEM results show that the biodegradable plastic is getting denser due to increasing chitosan concentration. The thermal analysis shows good thermal stability. The results of FTIR spectrum analysis on a mixture of glycerol starch and chitosan with wave numbers 1049-1662.64 contained C-C, C-N, C-O and C=C groups. Apart from that, the addition of glycerol affects water absorption capacity and biodegradability analysis occurs within 3 days.

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## Authors Contributions

Fadlan Hidayat and Rita Sunartaty conceptualized the research idea, designed of methodology, management and coordination responsibility; Ibrahim and Safiah analyzed data, conducted a research and investigation process; Liya Fitriyana conducted literature review and provided critical feedback on the manuscript.

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## Conflicts of Interest

The authors declared no conflict of interest.

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