

Producing starch-based bioplastic from durian skin with the addition of sorbitol plasticizer and chitosan filler

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Abstract

Plastic and organic waste are global issues in various developing countries, especially Indonesia. Improper waste management will bring a negative impact on the environment. Organic waste such as durian skin can be converted into starch as a raw material for bioplastics as an alternative solution to reduce plastic waste. This study aims to make bioplastics based on starch from durian skin with the addition of sorbitol as a plasticizer and chitosan as a filler with the addition of tapioca flour adhesive. The ratio of tapioca flour to durian skin starch is 1:1, 1:0.75, 1:0.5, while the concentration of sorbitol plasticizer used is 25 %, and chitosan filler is 4 % of the total starch mass. The samples were characterized by physical and mechanical tests (thickness, tensile strength, elongation, and young's modulus), functional group tests with FTIR, and thermal strength tests with TGA-DTG. From the results, the best sample was sample C (the ratio of tapioca flour to durian skin starch 2: 2, sorbitol plasticizer 3.00 ml, and chitosan filler 0.48 g) with a thickness of 0.11 mm, tensile strength 31.44 MPa, young's modulus 31.23 Mpa, and stable thermal resistance with the smallest mass loss of 81.454 % up to a temperature of 600 °C.

Keywords: bioplastic, chitosan filler, durian skin starch, sorbitol plasticizer

How to Cite: Rahmatullah, Putri, R. W., Sufra, R. (2024). Producing starch-based bioplastic from durian skin with the addition of sorbitol plasticizer and chitosan filler. *Jurnal Teknik Kimia*, 30(3), 164-179. <http://doi.org/10.36706/jtk.v30i3.2556>

1. INTRODUCTION

Plastic and organic waste or waste is a global issue in some developing countries, especially Indonesia. In South Sumatra, the waste reached 886,632 tons per year. The waste is divided into household waste 47.05 %, traditional markets 22.31 %, areas 9.53 %, business centers 8.22 %, offices 6.09 % and public facilities 5.36 %. This waste is dominated by food waste of 40.91%, and plastic waste 18.9%, the rest from leaves and twigs 13% and paper 10.63%. (Ministry of Environment and Forestry, Directorate General of Waste Management, Waste and B3 Directorate of Waste Management., 2023).

As one of the efforts to reduce the amount of waste, especially plastic and organic waste, is waste processing and management, including processing organic waste

such as durian skin waste into bioplastics. Durian skin has a crude fiber content (62.46 %), starch (36.8 %), protein (19.9 %) and lignin (10.53 %) (Haryati et al., 2018). This high starch content of durian skin potentially to use as a raw material for bioplastics.

Several studies on starch-based bioplastics have been widely developed. In 2020, Zalfiatri et al. conducted a study on the manufacture of biodegradable bioplastics from durian seed starch and jackfruit seed starch. The treatments in this study were P1 (5 % jackfruit seed starch), P2 (1.25 % durian seed starch: 3.75 % jackfruit seed starch), P3 (2.5 % durian seed starch: 2.5 % jackfruit seed starch), P4 (3.75 % durian seed starch: 1.25 % jackfruit seed starch), and P5 (5 % durian seed starch) in the biodegradable plastic formulation. From the results of this study, the best sample was P2 which had a thickness of 0.76 mm, plastic resistance to water of 59.19 %, water vapor transfer rate of 4.53 g/cm²/hour, tensile strength of 21.57 MPa, elongation of 6.58 % and plastic biodegradability for 2 weeks.

In the process of making starch-based bioplastics, the addition of plasticizers and fillers is required to improve the quality characteristics of bioplastics. Plasticizers are used as additives that increase the flexibility and durability of a material. Fillers function to strengthen or harden the material of a composite. Fillers work based on the principle of adhesion, which is the attractive force between molecules of different types of materials (Melani et al., 2017).

Fitriany et al., (2023) successfully made bio-pack: biodegradable cassava starch packaging as a solution to conventional plastic waste pollution using glycerol as a plasticizer and chitosan as a filler. In this study, variations 1 (glycerol 15 ml without chitosan), 2 (chitosan 20 ml without glycerol), and 3 (glycerol 20 ml and chitosan 15 ml) were used. From the results of this study, it is proven that the composition of bioplastic is more effective in variation 3 because it has a very strong structure and when tested for heat resistance, it is only slightly torn and has a soft texture, and the speed of change in the texture of the bioplastic runs normally for 3-5 minutes. From the degradation test, it was also proven that the sample with variation 3, the shape of the plastic after degradation for 7 days was damaged, and the time required for the degradation process was relatively more normal than variations 1 and 2.

In 2024, Enda et al. conducted an analysis of the use of plasticizers (glycerol and sorbitol) in the manufacture of bioplastics from yam starch using 5 % CH₃COOH solvent. The main qualitative research methods in this study were observation of test phenomena, biodegradation assessment, and assessment of water absorption capacity. Based on samples with glycerol sorbitol content of 15 %, 20 %, and 25 %. The 15 % concentration group experienced damage or weight loss faster than samples containing 20 %, 25 %, and 15 % glycerol. Water absorption is related to the ability of bioplastics to survive storage time. From the results of the analysis of biodegradation tests and water absorption, the lower the concentration of glycerol and sorbitol, the more fragile the plastic produced, less elastic, more easily decomposed by water, soil, and other microorganisms. The higher the concentration of glycerol and sorbitol, the higher the percentage of results obtained. The resulting plastic will be harder so that it is not easily decomposed or damaged.

Initial research on the manufacture of starch from durian skin has been conducted by Nurrohmah et al., (2021), where in this study the use of durian skin starch was limited to food ingredients, not as a raw material for bioplastics. Therefore, considering the potential of durian skin starch as a raw material and the important role of plasticizers and fillers in the manufacture of bioplastics, this study will focus on the manufacture of starch-based bioplastics from durian skin and how the addition of sorbitol plasticizer and chitose filler affects the resulting bioplastics.

2. MATERIALS AND METHODS

2.1 Tools And Materials

The raw materials used in this study were durian skin from the traditional market in the Kuto market in Palembang, commercial tapioca flour, sorbitol, chitosan, and distilled water. The tools used were aluminum foil, 100 mesh sieve, stirring rod, glass beaker, hot plate, magnetic stirrer, oven, analytical balance, dropper pipette, and thermometer.

2.2 Preparation of Raw Materials (Modification of nurrohmah's research, 2021)

Durian skin waste is prepared, then the white part of the durian skin is separated and cut into small pieces. Next, the durian skin is washed clean using running water, then dried in the sun for approximately 3 days until dry, then dried again using an oven to reduce the remaining water content at a temperature of 100 °C to a constant weight.

2.3 Research Procedure

The research procedure was based on research by Melani et al., (2017) with the following stages. Weight 6 grams of starch with a ratio of tapioca starch and durian skin starch (1:1; 1:0.75; 1:0.5). Sorbitol plasticizer and chitosan filler preparation, each as much as 25 % and 4 % of the total starch mass. The 25 % plasticizer from the total starch mass into a beaker containing 58 ml of distilled water. Add filler with a concentration of 4 % of the total starch mass. Do this for each chitosan filler. The last stage, add the starch mixture then stir and heat for 10 minutes at a temperature of 60 °C continuously. Repeat for each variation of the ratio of tapioca starch and durian skin starch.

Print the bioplastic solution using a petri dish. Then the bioplastic solution is dried in an oven at a temperature of 45 °C for 5 hours or constant weight. Remove the mold from the oven and dry it at room temperature until the bioplastic can be released from the mold.

Tabel 1. Research Matrix

Sample	Ratio of tapioca starch: durian skin starch	25 % sorbitol (ml)	4 % chitosan (g)
A	1:0.5	2.25	0.36
B	1:0.75	2.63	0.42
C	1:1	3.00	0.48

2.4 Analysis

The characteristics of bioplastics were tested using FTIR to determine the functional groups of each component, mechanical tests in the form of tensile strength and elongation, and thermal resistance tests using TGA-DTG.

3. RESULTS AND DISCUSSION

3.1 Durian skin starch bioplastic thickness test results

Thickness is an important parameter that affects the use of film in the formation of the product to be packaged. The thickness of the film will affect gas permeability. The thicker the plastic film, the smaller the gas permeability and better protect the packaged product (Putra et al., 2019). Thickness can also affect other mechanical properties of the film, such as tensile strength and elongation. However, in its use, the thickness of the film must be adjusted to the product being packaged (Nafilah & Sedyadi, 2019). The results of the thickness test of the bioplastic thickness from durian skin starch can be seen in Figure 1.

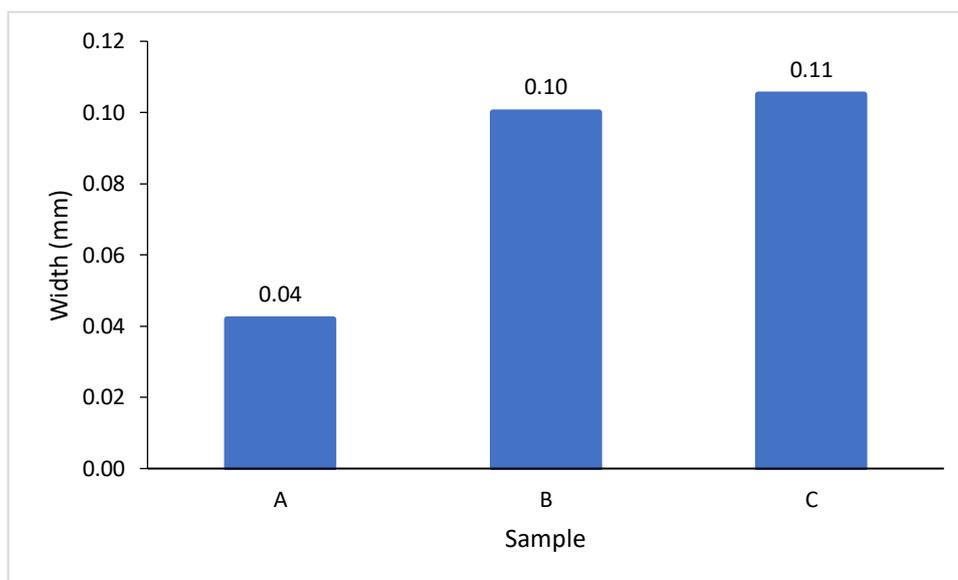


Figure 1. Durian skin starch bioplastic thickness test results

The treatment of adding starch concentration to the thickness of bioplastics produced an average thickness value ranging from 0.04 mm to 0.11 mm. The lowest bioplastic thickness was found in sample A with a ratio of tapioca flour starch: durian skin starch 1: 0.5, sorbitol plasticizer 2.25 ml, and chitosan filler 0.36 g of 0.04 mm, while the highest value was obtained in sample B with a ratio of tapioca flour starch: durian skin starch 2: 2, sorbitol plasticizer 3 ml, and chitosan filler 0.48 g of 0.11 mm.

The results of the various analyses show that the addition of durian peel starch concentration in the manufacture of bioplastics has a significant effect on the level of thickness produced. The low thickness of sample A bioplastic is influenced by the amount of solution solids used in the manufacture of bioplastics (Budiman et al., 2018). The amount of dissolved solids in sample A is lower than other treatments. This is supported by the statement of Imran et al., (2014), which states that the thickness of bioplastic is influenced by the amount of dissolved solids and the surface area of the container, different thickness values are caused by the amount of dissolved solids which are the components. Meanwhile, according to Apriyani & Sedyadi (2015), the thickness value increases with the increasing amount of starch used. The thickness of bioplastic is influenced by the total amount of solids in the solution and the size of the mold. The more total solids in the solution, the thicker the bioplastic produced in the same mold. Febiyanti et al., (2023) explain that the increasing concentration of the material used will increase the thickness of the plastic. The increase in thickness

occurs due to differences in the concentration of the plastic-making material, the container or mold used, while the volume of plastic solution poured into each plate is the same.

The thickness value of bioplastic in the study of making bioplastic from cassava waste starch, conducted by Apriyani & Sedyadi, (2015), with a starch mass concentration of 4-6 g produced the lowest thickness value of 0.05 mm and the highest thickness value of 0.08 mm. The addition of starch and plasticizer to bioplastics is directly proportional to their thickness. This is in line with the results of research conducted by Budiman et al. in 2018, bioplastics were made by varying lindur starch (0.5; 1; 1.5; and 2 gr), and glycerol plasticizer 15% of the total composite (starch mass and chitosan 4 gr). The results of this study showed that the highest thickness was obtained at a concentration of 2 gr starch, 4 gr chitosan and 15% glycerol (total composite) equivalent to 0.9 mL resulting in a thickness value of 0.11 mm.

3.2 Results of tensile strength test of durian skin starch bioplastic

Tensile strength is the maximum amount of force a material can withstand before breaking or deforming permanently when stretched or pulled. This test aims to determine the resistance of a material to loading at the bending point and also to determine the elasticity of a material (Soraya, 2020). Figure 2. shows the results of the tensile strength test of durian skin starch bioplastic.

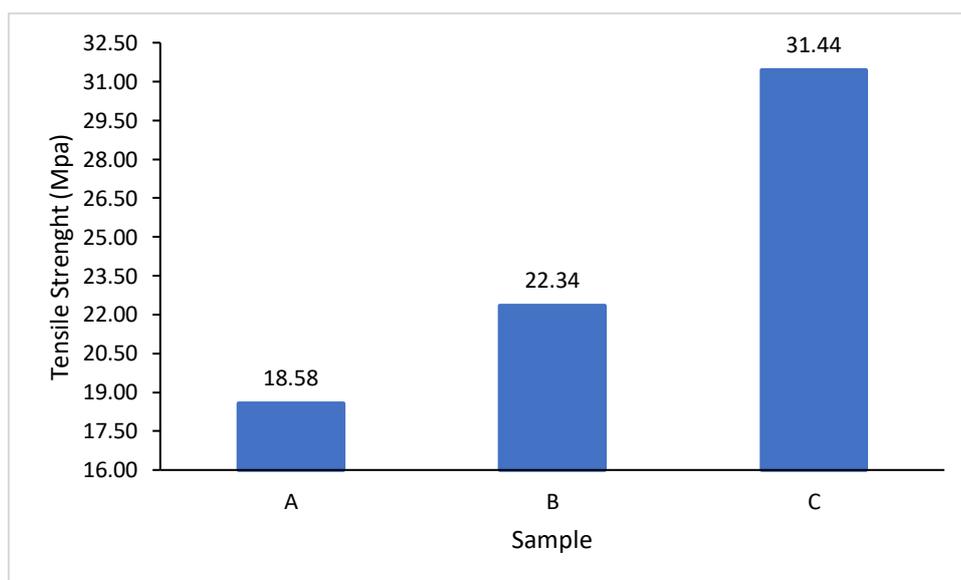


Figure 2. Results of tensile strength test of durian skin starch bioplastic

Figure 2 above shows that the addition of each component, namely the mass of durian skin starch, the concentration of sorbitol plasticizer and chitosan filler, can increase the tensile strength of bioplastics. The lowest tensile strength value was obtained in sample A with a ratio of tapioca flour: durian skin starch 1: 0.5, sorbitol plasticizer 2.25 ml, and chitosan filler 0.36 g of 18.58 MPa, while the highest value was obtained in sample B with a ratio of tapioca flour: durian skin starch 2:2, sorbitol plasticizer 3 ml, and chitosan filler 0.48 g of 31.44 MPa. The addition of starch to bioplastics will increase the amylose content in the solution so that it will cause the number of hydrogen bonds formed to increase, so that the bonds are difficult to break (Apriyani & Sedyadi, 2015). Strong hydrogen bonds between polymers will make the formed film hard and less flexible, and vice versa (Wirawan, 2013).

The addition of chitosan also affects the tensile strength value. Chitosan causes the formation of interactions with the cellulose polymer chain in the form of hydrogen bonds, where the interaction of this polymer chain is formed to increase the speed of viscoelastic response in the polymer so that it can increase the mobility of the polymer chain (Pariwi et.al., 2016). Tensile strength increases with the increasing starch weight ratio due to the formation of the intermolecular hydrogen bonds between NH^{3+} cations of the chitosan backbone and OH^- anions of the starch. (Navarro, et al. 2019) The addition of sorbitol can also increase the tensile strength value of plastic, but with excessive composition the tensile strength value can decrease. This is in line with research conducted by Yustisi et al., (2024), that the addition of sorbitol can also increase the tensile strength value of plastic, but with excessive composition the tensile strength value can decrease. In this study, the maximum tensile strength value was obtained in bioplastic with a starch composition of 4 grams with the addition of 5 grams of chitosan and 6 ml of sorbitol, namely 49.94 Mpa.

According to the Japanese Industrial Standard (JIS), the minimum standard value of tensile strength for plastic food packaging is 3.92266 MPa (Ariska & Suyatno, 2015), while according to the Indonesian National Standard (SNI), the tensile strength standard ranges from 24.7-302 MPa (Adil et al., 2020). The tensile strength values obtained in the study have met the plastic standards set by both JIS for samples A (18.58 MPa) and B (22.34 MPa) as well as sample C (31.44 MPa) for the SNI standard.

3.3 Results of elongation test of durian skin starch bioplastic

Percent elongation is the change in the length of the bioplastic after the bioplastic breaks when stretched. Based on Figure 3. the percent elongation value obtained is still fluctuating. The percent elongation value increases with the addition of a starch ratio of 1:0.75, 2.63 ml plasticizer, and 0.42 g chitosan filler from 0.68 % to 1.81 %, then decreases with the addition of a starch ratio of 1:1, 3 ml plasticizer, and 0.48 g chitosan filler to 1.01 %.

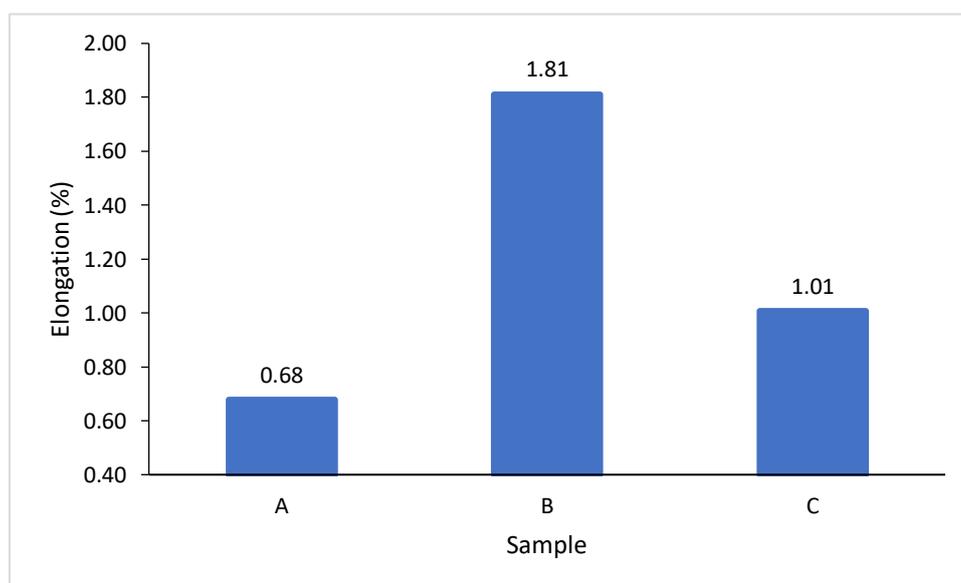


Figure 3. Durian skin starch bioplastic elongation test results

The elongation value will affect the quality of bioplastics, where a higher elongation value indicates that the bioplastic has good mechanical properties. The greater the addition of plasticizer, the percentage of elongation will increase, but after the addition

reached at a certain concentration the value will decrease, because the more concentration of plasticizer the cohesion bonds between polymers will be smaller and the film formed will be softer so that the edible film formed is easily broken (Bourtoom, 2008). The stronger the breaking distance of the bioplastic film causes the film to have brittle and easily brittle properties, so that the elongation value decreases (Nurhaliza et al., 2022).

Based on research by Ramadhani et al. (2022), Lusiana et al., (2019), and Oluwasina et al., (2019), which states that along with the addition of plasticizer concentration to bioplastics, the percentage elongation value will also increase. However, adding sorbitol after reaching its critical point will cause a decrease in the elongation value (Nafilah & Sedyadi, 2019). In several study, too much sorbitol plasticizer was used, the elongation value decreased. (Harumarani & Ma'ruf, 2016; Khotimah & Tjahjani, 2020; Mappamadeng & Amalia, 2021).

According to research conducted by Hartatik & Nuriyah, (2014), the addition of chitosan concentration in a certain amount also causes a decrease in the percentage of elongation in bioplastics. The results of testing durian skin starch bioplastics showed that the elongation value did not meet SNI standards with a range of values of 21.00-220.00 %.

3.4 Results of the Young's modulus test of durian skin starch bioplastic

Young's modulus can be said to be a measure of the stiffness of a material or the difficulty of the material to deform when subjected to a load (Zherebtsov et al., 2019). The value of the young modulus is obtained by calculating the comparison between tensile strength and elongation at break (Rifaldi et al., 2017). The results of the young modulus of durian skin starch bioplastic are presented in Figure 4.

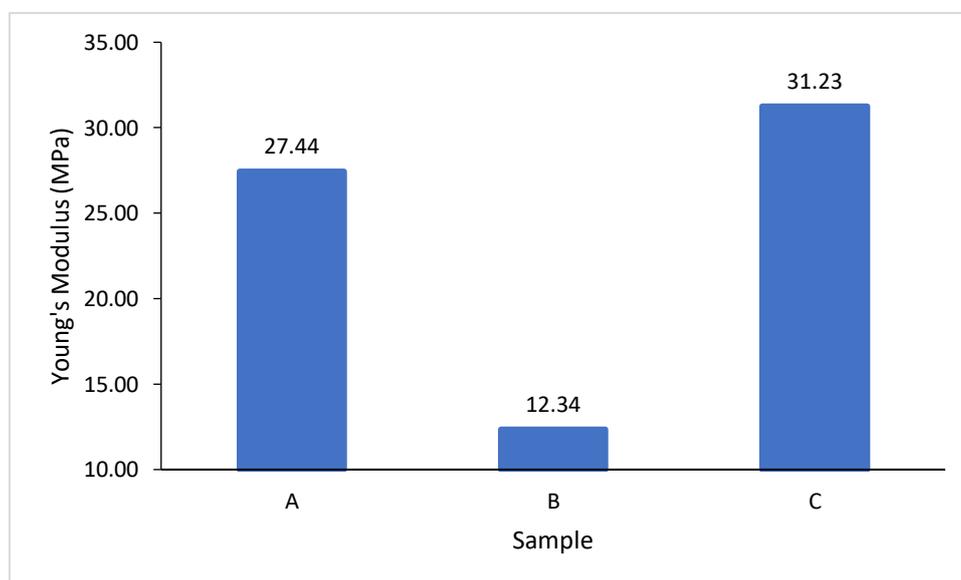


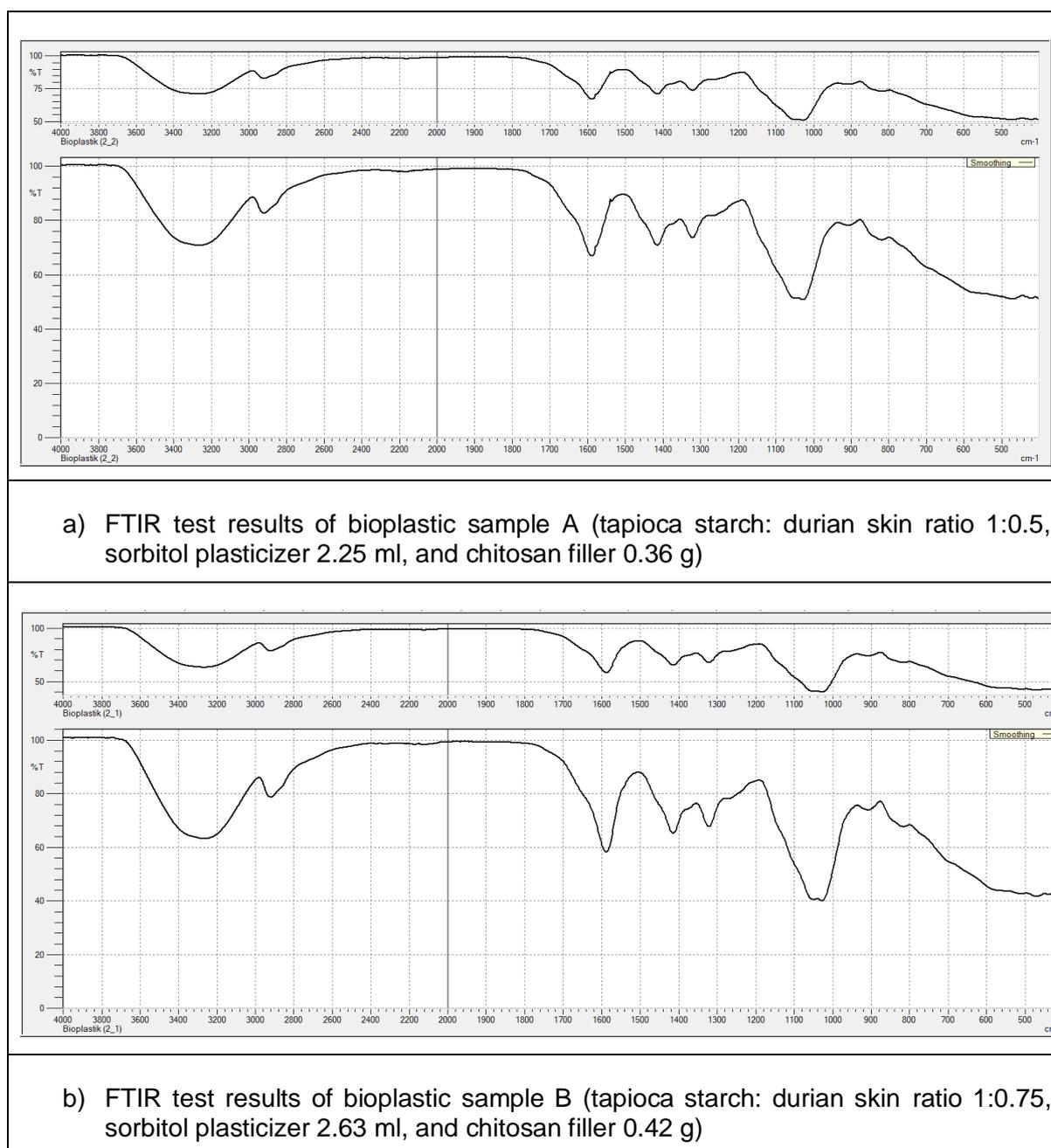
Figure 4. Young's modulus test results for durian skin starch bioplastic

In Figure 4, the highest Young's modulus value was obtained in sample C with a starch ratio of 1:1, sorbitol plasticizer 3 ml, and chitosan filler 0.48 g of 31.23 MPa. The higher the Young's Modulus value, the more difficult a bioplastic is to bend or stretch and the higher the stiffness of the material. If the Young's modulus value is too high,

the bioplastic will also lose its elasticity (Yustisi et al., 2024). Figure 4 shows a decrease in the Young's Modulus value with a starch ratio of 1:0.75 with a variation of 25% sorbitol and 4% chitosan (Sample B), then an increase occurs at a starch ratio of 1:1 with the same variation to the total mass of starch (Sample C). This can occur because the tensile strength value is high and the addition of a lot of sorbitol causes the elongation value to be low, so the Young's Modulus value is high. (Yustisi et al., 2024).

3.5 FTIR test results of durian skin starch bioplastic

The results of the FTIR analysis for each sample of durian skin starch-based bioplastic can be seen in Figure 5 and Table 2 below:



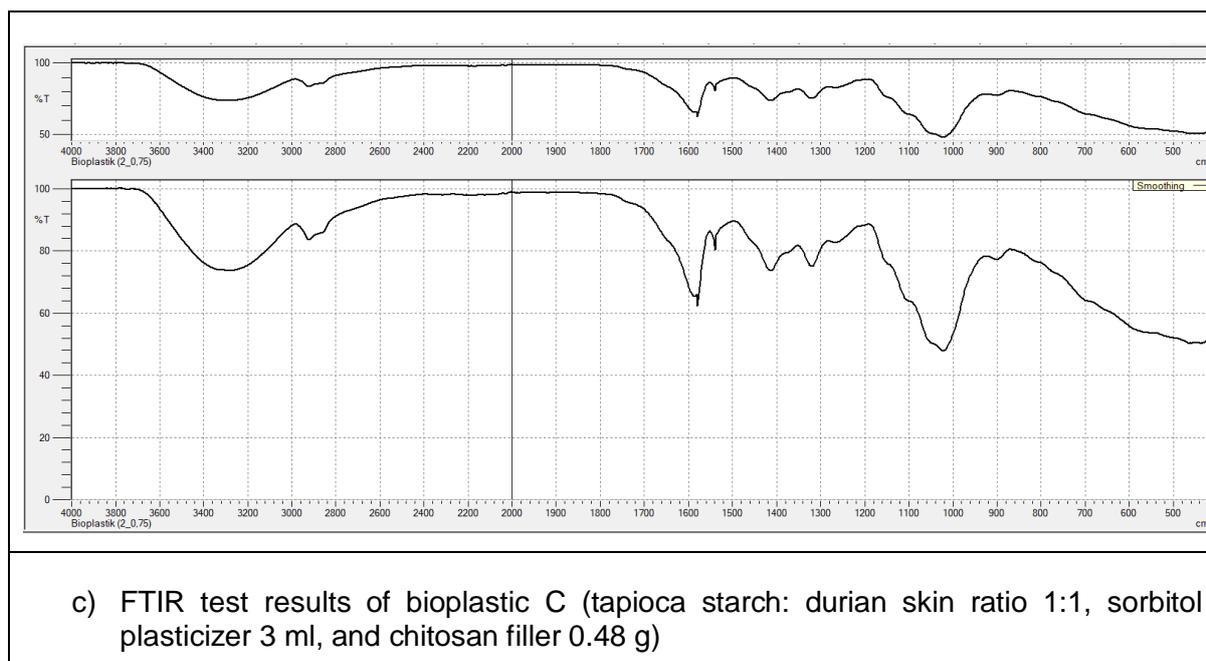


Figure 5. FTIR results graph of bioplastic samples

Table 2. Results of FTIR Analysis on The Main Groups of Bioplastic Samples and The Comparison on Their Constituent

Component	Wavenumber (cm ⁻¹)							
	-OH	-CH	C=O/-CN amida	NH amina	CH amina	-OH/NH ₂	C-O	C-C
Starch*	3388	2929						1649
Sorbitol**	3257	2937						
Chitosan***		2877.79	1658.79	1597.06	1381.03	3441.01	1033.85	894.97
A	3252.60 3370.30	2888.01 2988.49	1653.57	1588.98	1379.41	3443.50	1030.61	895.68 1647.83
B	3262.65 3351.64	2635.38	1653.57	1588.98	1382.28	3351.64	1034.92	895.68 1647.83
C	3251.16 3386.09	2721.50 2981.31	1653.57	1588.98	1375.11	3463.60	1034.92	894.25 1647.83

(*,**Rahmatullah et al., 2022; ***Saputro & Ovita, 2017)

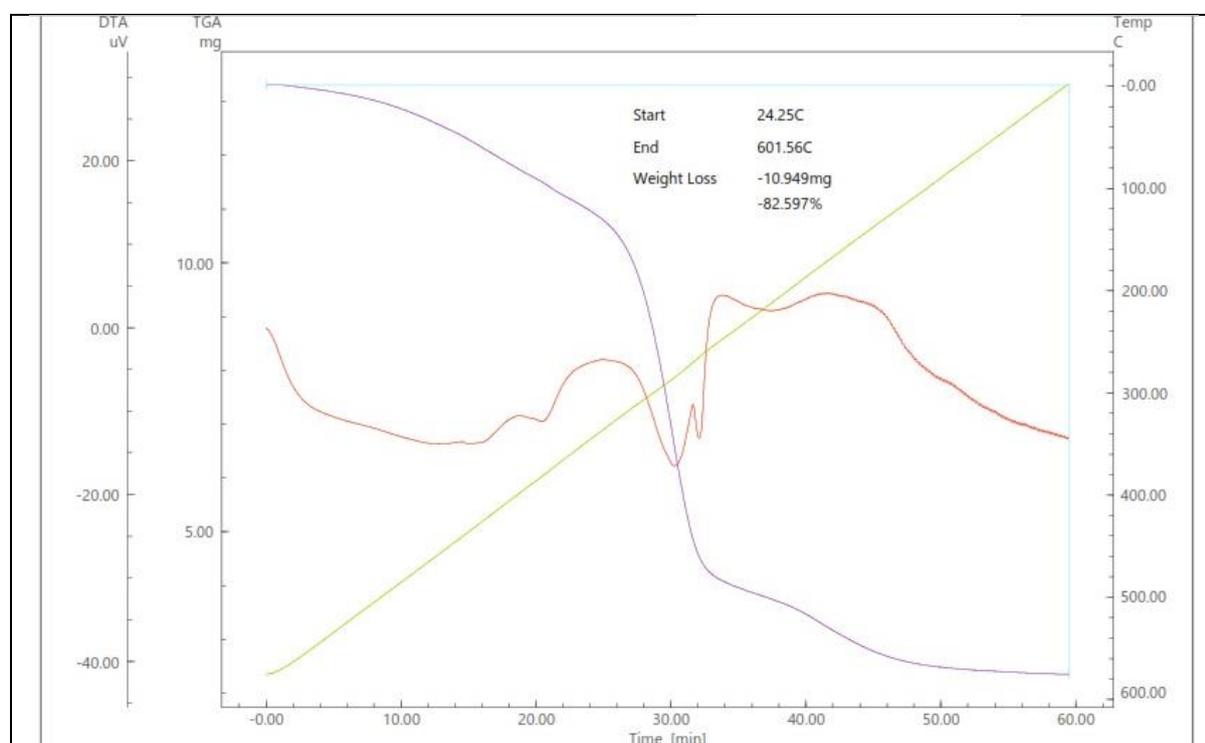
Based on Figure 5 and Table 2, it can be seen that the resulting bioplastic has a group that is relatively the same as its constituent components, the bioplastic formed still has hydrophilic properties with the presence of OH- groups originating from sorbiol and starch with wavelengths of 3370.30 and 3252.60 respectively; 3351.64 and 3262.65; 3386.09 and 3251.16 for samples A, B, and C respectively. The presence of this hydroxyl group indicates the presence of a polymer compound formed. The presence of the C-C group at a wavelength of 1647.83 indicates the starch content in the bioplastic (Rahmatullah et al., 2022).

The presence of a strong and wide absorption band with a wavelength area in the range of 3400 cm⁻¹ indicates the presence of -OH groups that overlap with -NH₂. The widening of the spectrum area at a wavelength of 3400 - 3463 cm⁻¹ is caused by the addition of -OH groups due to the addition of starch to each sample. Based on Figure 4 and Table 1, in the FTIR spectra of durian skin starch bioplastic, no new functional group peaks appear, all wavelength peaks that appear are groups from each component of the bioplastic. This shows

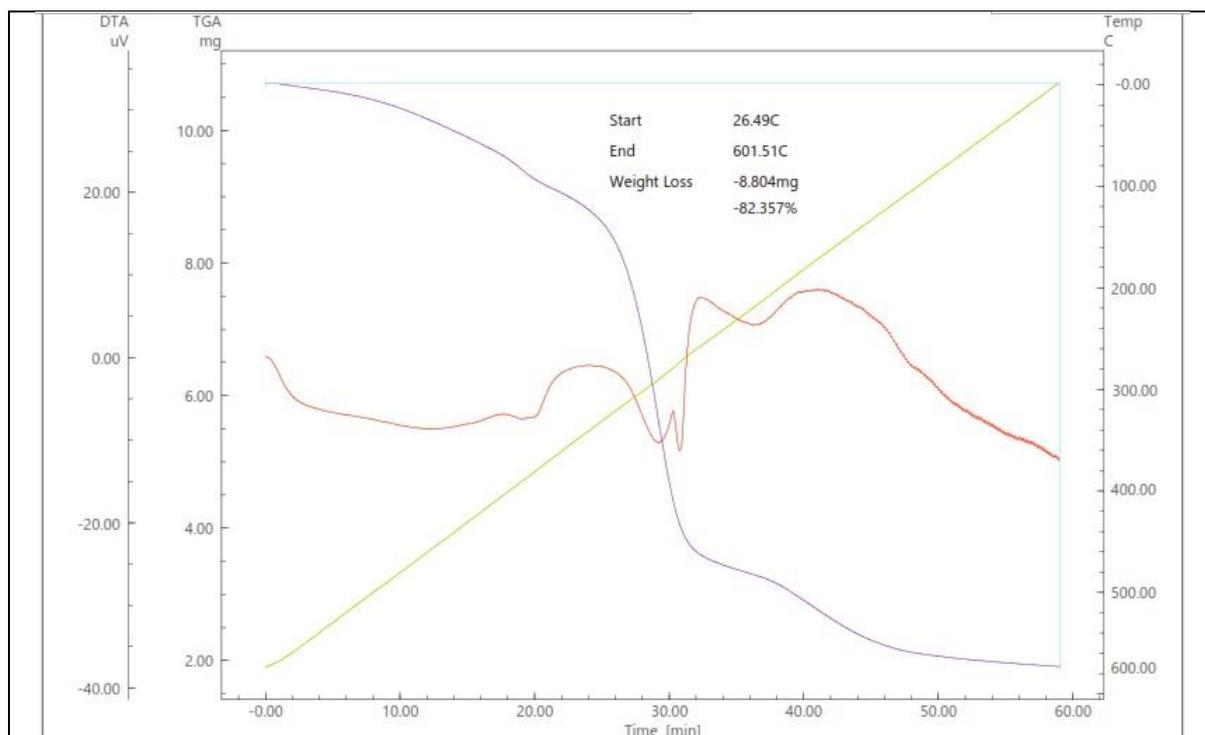
that the bioplastic formed is the result of physical mixing and no reaction is formed (Saputro & Ovita, 2017).

3.6 Results of thermal analysis test of durian skin starch bioplastic

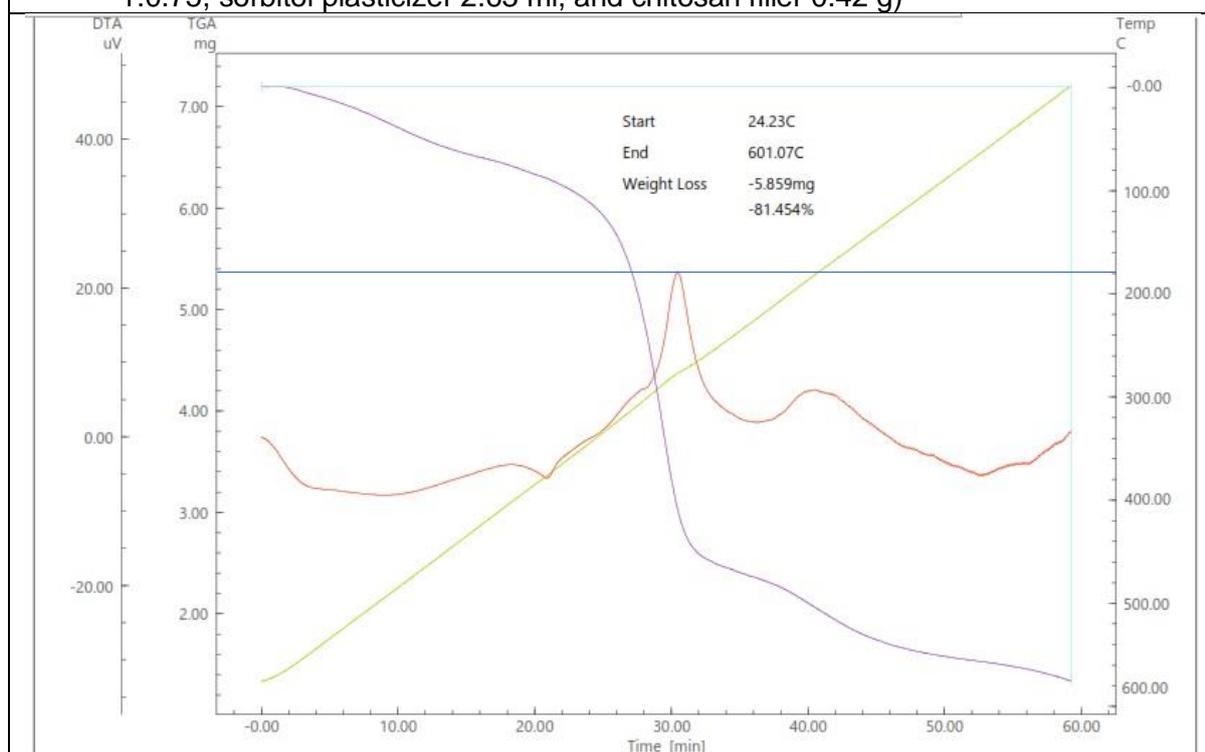
Thermal property characterization can be measured using a tool called Thermogravimetric Analyzer (TGA) and Differential Thermal Analysis (DTA) to show the nature of a material. Thermal processes include phase change, softening, melting, decomposition and oxidation. Thermal property analysis includes measuring the melting point (T_m), glass transition temperature (T_g), and enthalpy changes of the sample during the process (Waldi, 2007). The results of thermal analysis are in the form of a curve called a thermogram. Thermograms can show endothermic and exothermic enthalpy changes. The results of this thermal analysis in the form of a thermogram are intended to determine the thermal stability of environmentally friendly plastic made from durian skin starch. From the results of this analysis, information was obtained about the physical changes in bioplastics shown in Figure 6.



a) TGA-DTA test results of bioplastic sample A (tapioca starch: durian skin ratio 1:0.5, sorbitol plasticizer 2.25 ml, and chitosan filler 0.36 g)



b) TGA-DTA test results of bioplastic sample B (tapioca starch: durian skin ratio 1:0.75, sorbitol plasticizer 2.63 ml, and chitosan filler 0.42 g)



c) TGA-DTA test results of bioplastic sample C (tapioca starch: durian skin ratio 1:1, sorbitol plasticizer 3 ml, and chitosan filler 0.48 g)

Figure 6. TGA-DTG results graph of bioplastic form durian skin stretch

The results of the thermal analysis test can be seen in Figure 6, thermal analysis was carried out simultaneously starting from a temperature of 24-601.5 °C.

The results of the TGA-DTG analysis, TGA on the purple line, and DTA on the red line obtained results with a gradual decrease in the weight of the bioplastic. Based on Figure 4. The TGA thermogram provides information about the decrease in mass due to an increase in temperature while the heat seen in each mass change process can be seen from the DTA thermogram. The TGA thermogram data in Figure 4. shows that each sample experienced a drastic decrease in mass on average in the same temperature range starting from 110 to 460 °C, then decreased again to a temperature of 600 °C. The process of reducing the mass (thermal degradation) of bioplastics takes place in 2 stages. In the first stage, a temperature of 110 °C is the loss of water bonds from the sample. The initial mass reduction in bioplastics occurs due to the loss of water contained in the bioplastic due to evaporation (Setiawan et al., 2021). The second stage, temperature 460 °C is caused by thermal decomposition of molecules and samples consisting of small molecules of carbon and hydrocarbons from semi-crystalline starch components along with sorbitol and chitosan. The results of this study are the same as those conducted by Puspita, (2013), where the starch/PVA film showed two main stages of degradation. The first degradation of water loss in the sample occurred at 209.1 °C. The second degradation was around 314.5 °C and it was caused by thermal degradation of semi-crystalline starch. Almost 50 % of the total film was degraded at 369 °C. The starch/PVA mixture film lost mass reaching 90 % at 423.5 °C.

At the final degradation temperature of 600 °C. Sample A with an initial weight of 13.256 experienced a mass reduction of 10.949 mg or 82.597 % of the total mass lost, sample B with an initial weight of 10.690 experienced a mass loss of 8.804 mg or 82.357 %, while sample C with an initial weight of 7.193 mg had a total mass reduction of 5.859 mg or 81.454 %.

At a temperature of 280 °C, the main stage of degradation that occurs in bioplastics is the glass transition where solid and rigid bioplastics will change form to soft and elastic, this change is influenced by high temperatures. At a temperature of 300 °C, depolymerization occurs, namely the process of breaking the polymer chain in bioplastics, thereby reducing the weight of the bioplastic. At a temperature of 370-600 °C, the melting point occurs where solid bioplastics will change form to liquid, so it can be said that the resulting bioplastic can be used as packaging and can withstand heat up to 210 °C. However, at temperatures above 210 °C, bioplastics will degrade (Fatwa et al., 2022).

The mass reduction process for samples A and B shows an exothermic process (temperature increase) with a melting point of 360 °C on the DTA thermogram, this peak illustrates the temperature at which the material begins to burn or decompose. In sample C, the opposite occurs, there is a decrease in temperature at a temperature of 220 °C which indicates that the process occurs endothermically. Identical to the melting point of synthetic plastic film type PVA (polyvinyl alcohol) where the endothermic peak also occurs at 222 °C. This peak is identified as the temperature at which the material begins to burn. At that point, the bioplastic loses moisture in its structure (Puspita, 2013). This material does not melt but burns immediately, this is because the material contains components as a strengthener or filler. The endothermic process is a process of water removal that occurs during the heating time (Salbeti et al., 2018). Based on the results of the thermal test on the TGA-DTA thermogram, it shows that the variation that is more thermally stable is in sample C when viewed from the smallest mass reduction of 81.454 %.

Several similar research results have been reported on the thermal resistance of bioplastic materials. Agustin et al., (2014) reported that the thermal resistance test

of bioplastics from starch and straw with the condition of the mass ratio of starch to nanocrystalline cellulose resulted in a mass reduction of 79.63 % at a temperature of 600 °C. In 2021, Setiawan et al., also showed similar results for bioplastic samples with a mixture of starch and rice straw cellulose, there was a mass reduction of 81.01 % from the initial mass at a temperature of 550 °C.

4. CONCLUSION

The manufacture of bioplastics with starch as the base material from durian skin flour has been successfully carried out. Based on the results of the FTIR functional group test, no new compounds were found, or it can be interpreted that the bioplastic formed is the result of physical mixing and no reaction is formed. From each variation carried out, optimum results were obtained in sample C (the ratio of tapioca flour to durian skin flour starch 2:2, sorbitol plasticizer 3.00 ml, and chitosan filler 0.48 g) with a thickness of 0.11 mm, tensile strength 31.44 Mpa (meets the Indonesian national standard for bioplastics in the range of 24.7-302 Mpa), young modulus 31.23 MPa, and thermally stable up to a temperature of 600 °C, with the smallest mass reduction of 81.454 %.

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