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Potency of Utilizing Sago Starch as Natural Resource from Papua in the Production of Biodegradable Plastic

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***Abstract*— The use of conventional plastic has become a priority in various aspects of life but has contributed to environmental problems from the aspect of managing plastic waste. In this study, the feedstock of Sago Starch from Papua was utilized for its potency to produce biodegradable plastic by varying several parameters of chitosan, glycerol, and acetic acid at different levels. The optimum biodegradable plastic characterization of tensile strength of 7.41 MPa, elongation at break corresponding to 24.17%, and the fastest of 40 days durability of completely removed wastes of degradable plastics were achieved. Based on the results obtained, it was concluded that the strength of biodegradable plastic was affected by the amount of chitosan added, while the level of breaking downlinked to elongation at break and degradable period was related to the amount of glycerol as plasticizer used. Acetic acid was found to improve the solubility of the starch by acting as a catalysator, therefore making the homogenization become easily achieved. In addition, the SAS model was used to perform the interaction of all parameters to the characterizations measured of tensile strength and elongation at break. Hence, statistically, all glycerol, chitosan, and acetic acid parameters significantly affected biodegradable plastics characterizations ($p < 0.05$). By utilizing the right production technology through the stages of feedstock preparation, heating, chemical mixing, and printing, it is expected to produce good quality of biodegradable plastic, eco-friendly product and feasible to support the development of the economic sector in Papua.**

***Keywords*— Sago; biodegradable plastic; characterization; eco-friendly product; Papua.**

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I. INTRODUCTION

Biodegradable plastic is an alternative eco-friendly packaging product that has been intensively developed over the last two decades. Bioplastic is degradable material that pose no harm to the environment [1]–[4]. Biodegradable plastic is formed from biopolymer; a polymer derived from natural, renewable resources and crude oil [5]–[8]. One of the renewable sources for plastic production that is accessible globally and widely available in Indonesia is starch [1]–[4]. Starch is obtained by extracting vegetable substances that contain carbohydrates, such as corn, Sago, cassava, rice straw, sweet potatoes, sorghum, taro, and pineapple skin waste [4], [7], [9]–[12].

Biodegradable plastic manufacturing technologies are being developed worldwide, reviewed by several groups [3], [13]. However, in Indonesia, starch-based biodegradable plastic manufacturing technology has recently developed. When considering to develop starch-based bioplastic, several parameters to take into account may include a plasticizer, chitosan, acid as a catalysator, reaction time and temperature [3], [7], [8], [14]. Types of plasticizers that are commonly used are glycerol, gelatin, sorbitol. Chitosan can be extracted from crab and oyster shells. Acid was used in many studies to enhance plastic's transparency [15], [16]. In order to maintain strong mechanical properties while considering the softened structure of bioplastic, it is crucial to conduct the proper study.

One of the potential raw sources for starch is Sago. However, sago starch has been dominantly processed into food products despite its high potential to be used as biodegradable plastic raw material. Sago has unique functional characteristics which allow the starch to be examined for various purposes, both as food and non-food [4]. The Indonesian sago areas across the country are estimated at 1 million hectares. Meanwhile, the national sago starch potential is estimated to reach 2.5 million tons per year and has not been used optimally.

Specifically, in Papua, based on the previously reported data, the total area of sago palm forest was approximately around 1.200.000 hectares, while only 12.716 hectares were cultivated, corresponding to 4,763 kg/ha. Economically, if it is produced optimally, the amount of sago starch will reach 60.56 tons. However, based on the data obtained, only 42.79 tons of Sago had been collected in the farm, corresponding to 70% successfully harvested proceed. This fact indicated the lack of harvesting technology [17], [18]. Thus, several agricultural technologies need to be considered to expand the use of land and improve productivity in general [19].

In order to optimize the use of Sago in the non-food industry, this material has to be utilized as a feedstock for the production of biodegradable plastic as one of the derivatives products of Sago [20]. The purposes of this work were to determine which parameters significantly affect the characterization and the degradation rate of biodegradable plastic produced. Hence, this work would be very useful as a pioneer in producing biodegradable plastic in Papua Province and can potentially lead to industrial-scale production of Sago-based biodegradable plastics to support government regulation of limiting the use of conventional plastic in the province.

II. Materials and Methods

A. Materials

The material used in this study was Sago starch from a local farm in Jayapura, Papua. This feedstock was obtained and prepared for further characterization and biodegradable plastic production. Wet Sago was transferred into a sealed container to prepare for chemical characterization to obtain several composition properties. Meanwhile, feedstock was dried manually in an open area exposed to direct sunlight for 8 hours daily for three days to achieve a moisture content of less than 5% to produce biodegradable plastic.

Several chemicals applied were acetic acid (CH_3COOH) 5%, chitosan purity of 98% with particle size between 100 – 300 Mesh ordered from a company, and glycerol 85 % analytical standard as a plasticizer. In addition, distilled water and two different colorings used were red and green dyes. Tools used were hot plate magnetic stirrer (SH-2 Digital Lab Thermostatic Hot Plate), analytical balance Matrix Type AJ1002B, aluminum plates, glass beakers, spatula, aluminum foil, and glass plate, while instrument applied were drying oven and tensile strength tester.

B. Methods of Film Production

In this study, the composition of Sago starch prepared was 4 g, and distilled water was set to 50 ml for all twelve samples of experiments, while chitosan and glycerol composition were varied. A set of experiments is performed with a total of six combination parameters done without the addition of acetic acid CH_3COOH (AA). The design experiment was run using chitosan with three different levels of 0 g, 1 g, and 2 g, while glycerol of 2.5 ml and 5 ml was added differently. Another set of experiments was performed by adding 2 ml of acetic acid into each sample, as presented in Table 1. The mixture of each set was performed at temperatures between 75-85 ^{0}C during reaction time for 20 minutes. Although it is not necessary, this study was also aimed to see the possibility of colored products. Therefore, the coloring process was included. This step took place 1 minute before the reaction was completed by adding 1 ml of red or green dyes of food coloring into the mixture of starch, distilled water, with or without acetic acid while still at the hot plate. The steps of biodegradable plastic production from Sago starch can be seen in Fig. 1.

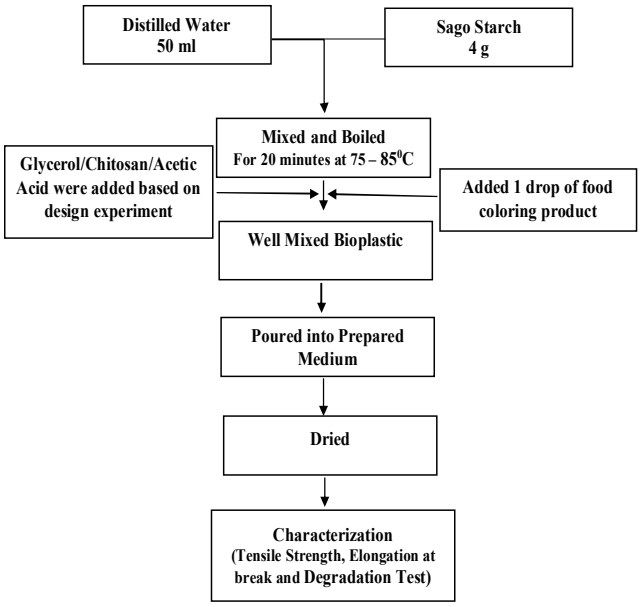

Fig. 1 Production Process of Biodegradable Plastic from Sago Starch

TABLE I
COMPOSITION PARAMETERS OF BIODEGRADABLE PLASTIC

S	WoS (gr)	DW (ml)	AA (ml)	C(gr)	G(ml)
S1	4	50	0	0	2.5
S2	4	50	0	1	2.5
S3	4	50	0	2	2.5
S4	4	50	2	0	2.5
S5	4	50	2	1	2.5
S6	4	50	2	2	2.5
S7	4	50	0	0	5
S8	4	50	0	1	5
S9	4	50	0	2	5
S10	4	50	2	0	5
S11	4	50	2	1	5
S12	4	50	2	2	5

Note: S (Sample), WoS (Weight of Sample), DW (Distilled Water); AA (Acetic Acid); C (Chitosan; and G (Glycerol)

After the reaction was performed, a biodegradable plastic film was transferred into an aluminum foil for drying at room temperature overnight, followed by drying at an open area facing the sunlight for 10 hours daily from 3 up to 5 days. When the biodegradable plastic film was dried, the next step was separated from the medium and stored for further necessary characterization.

C. Composition of Raw Wet Sago Starch

Several parameters of Wet Sago Starch have been measured at Balai Besar Industry Agro Bogor, with the results presented in Table 2 below. It is clearly shown that Raw Sago was originally in an acid condition corresponding to a pH of 3.44 and moisture content of 45%. Including Fat and some other important values measured, amylose and amylopectin consisted in Sago, making this feedstock theoretically feasible

to produce biodegradable plastic. All the tests performed were followed the procedures stated in this study.

TABLE II
PHYSICAL AND CHEMICAL COMPOSITION OF RAW WET SAGO STARCH

Parameters	Units	Results	Methods
pH		3.44	SNI 01-2891-1992-point 16
Moisture	%	45.0	SNI 01-2891-1992 point 5.1
Dush	%	0.03	SNI 01-2891-1992 point 6.1
Protein (Nx6.2)	%	0.06	SNI 01-2891-1992 point 7.1
Fat	%	0.19	SNI 01-2891-1992 point 8.2
Amylose	%	24.8	SNI 01-4447-1998 point 6.11
Amylopectin	%	33.9	IK 7,2,3

D. Characterization Tests of Plastics Based Sago Starch

Several tests were performed to ensure the feasibility of Sago starch into the production of plastic, such as Tensile strength, Elongation at Break, and Soil Burial. These characterization performances were done in two different Laboratories of the Physics Department and Initial Laboratory of Integrated Science and Technology. Specifically, tensile strength dan elongation at break was done by adapted methods from ASTM D 882. The samples were prepared by conditioning at a temperature between (23 ±2 ^{0}C) for 24 hours. In the process, samples were cut into smaller pieces to fit the instrument, followed by clamping both ends on a tensile testing machine. When the power is turned on, the force will be selected to accommodate measuring the length after the break. Meanwhile, elongation at break can be calculated when the original length and after the break were measured. This study was also carried out as a test for soil burial performance. The samples were placed into soil and measured for weight or mass loss every ten days by measuring the total weight of samples and tray using the equation below.

$$\text{Mass loss } (\%) = \left(\frac{Initial\ Mass - Final\ Mass}{Initial\ Mass}\right) x\ 100 \qquad (1)$$

As the amount of soil moisture for all trays was already measured, it is possible to calculate the weight of biodegradable plastic left in the tray.

E. Statistical Analysis

This study used a model to analyze individual and interaction effects to the dependent variables of tensile strength and elongation at break for all treatment combinations using SAS 9.3 (Cary, NC) within a 95% confidential limit. In this design experiment, three parameters or factors were used defined as classes of glycerol, chitosan, and acetic acid used, and each class has different levels, as seen from table 1. Thus, a PROC GLM model was adapted to find the significant effects from all combinations.

III. RESULTS AND DISCUSSIONS

Based on the research conducted, it was found that biodegradable plastic films were successfully prepared using variations composition of chitosan, glycerol as a plasticizer, and acetic acid. Several characterizations analyzed were tensile strength, elongation at break, and biodegradable performance described in this section.

A. Tensile Strength

In this particular test, the tensile strength of biodegradable plastic measured was affected mainly by the amount of glycerol and chitosan added. In comparison, the addition of acetic acid was observed to affect hydrolyzing of starch. Therefore, improved the solubility of Sago Starch during the production of biodegradable plastic and at the same time improved the transparency. The finding was proven statistically as each factor was found to significantly affect the dependent variable of the tensile strength ($p < 0.05$). The chitosan added particularly for three different levels of 0 g, 1 g, and 2 g resulted in the thickness and stiffness of biodegradable plastic produced. As the chitosan amount was increased proportionally, a higher tensile strength was observed for all samples.

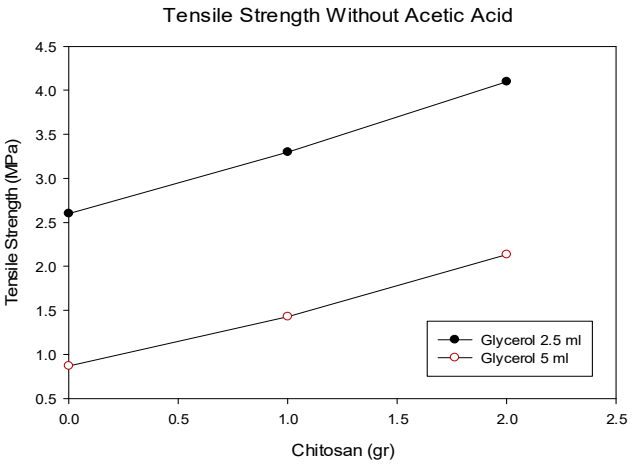

Fig. 2.a Tensile Strength Without AA

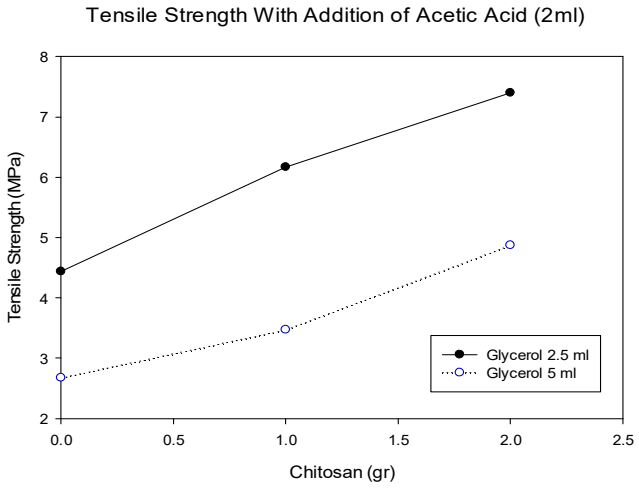

Fig. 2.b Tensile Strength With AA

As a result, biodegradable plastic was improved mechanically or structurally. Moreover, as the data gained, tensile strengths were found varied from 0.87 – 4.13 MPa without the presence of acetic acid as seen in Fig. 2.a and gradually increased to values of 2.63 – 7.41 MPa when acetic acid was added as presented from Fig. 2.b. In addition, the more glycerol presence in the solution mixture, the less tensile strength of plastic was achieved. The same phenomenon obtained the value of tensile strength when glycerol as plasticizer added was also done previously using Sago starch to produce Biodegradable plastic corresponding to the results were far lowered than our group [11], [21]–[23].

B. *Elongation at Break*

Chitosan is usually used as a starch mixture in the manufacture of biodegradable plastics to improve the mechanical properties of the biodegradable plastic produced [15], [24]. Data obtained observed that elongation at break tended to increase when the higher amount of glycerol was added, while it happened to decrease when a lower volume of glycerol was applied to correspond to volumes of 5.0 ml and 2.5 ml, as seen in Fig.3a and Fig.3.b. Specifically, when 5.0 ml of glycerol was used as reported from Fig.3.a below, without the addition of acetic acid, the optimum value of elongation at break reached was 17.06%. Meanwhile, when acetic acid of 2 ml was added, the maximum elongation at break was 24.17%, as shown in Fig.3. b. These two values correspond to a mild amount of chitosan used corresponding to 1 g.

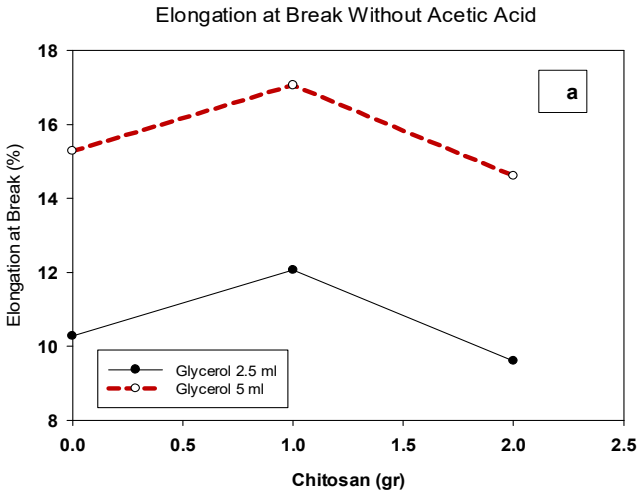

Fig. 3.a elongation at break Without AA

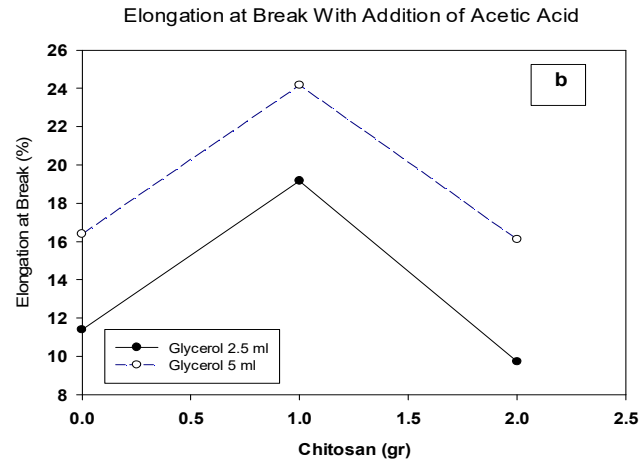

Fig. 3.b Elongation at break with AA

The findings from this study tended to support that plasticizer is an organic material with a low molecular weight that can reduce stiffness and increase polymer flexibility [15], [25], [26]. While statistically, all the factors of glycerol, chitosan, and acetic acid were found to affect the elongation at break for all compositions performed ($p < 0.05$). The measurement done in this study was also in line and consistent with the former findings, which reported the highest elongation at break observed to increase when the amount of glycerol used was improved [11], [27]. Moreover, one finding reported the value of elongation at break when Corm starch used as biodegradable plastic feedstock was increased as the plasticizer of Sorbitol used corresponding to value of 47.69% [23]. Moreover, other groups suggested there was a possibility to improve the elongation at break when different plasticizer used [4], [25].

C. *Degradation Test of Biodegradable Plastic*

Biodegradable plastic are easily breaking down into organic matters due to the interaction with environment [25], [28]. Particularly, the method used to ensure degradation rate was done by planting twelve different variations of parameters samples in the soil for days while observation and measurement were performed. The analysis was performed through visual observation on daily basis, while weighing each of plastic sample performed every 10 days. On the first day, all twelve samples appeared to be intact without having texture damage and becoming softer later as presented from Fig. 4.

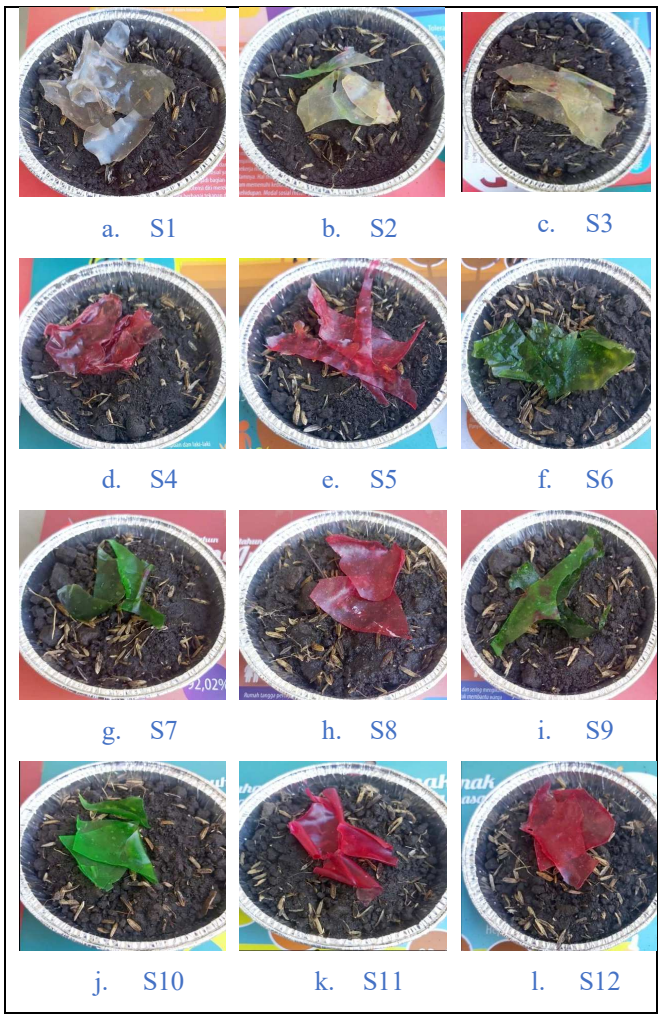

Fig. 4 First day observation of biodegradable plastic planted into soil

The highest degradable samples corresponding to 40 days of reaction were observed from samples S4 and S10 as seen from Fig. 4.d and Fig. 4.j. The results suggested that a lower amount of chitosan with acetic acid affected the mechanical strength to degrade faster. Meanwhile, the longest degradable

samples were achieved by samples named S3 and S9, as shown in Fig 4.c and Fig 4.i. Hence, the more chitosan means, the longer time needed to degrade completely. Thus, the findings were consistent with the previous group reported that chitosan enhances biodegradable plastic properties [24]. The same portion of time to degrade plastic was also observed from one previous work stating that bioplastic produced from banana corn starch took up to 77 days to degrade completely when a high amount of chitosan was used in the production [23]. As mentioned above, the duration was set up to 75 days for visual observation for all twelve samples. However, the measurement for the weight loss was done for the duration of up to 50 days.

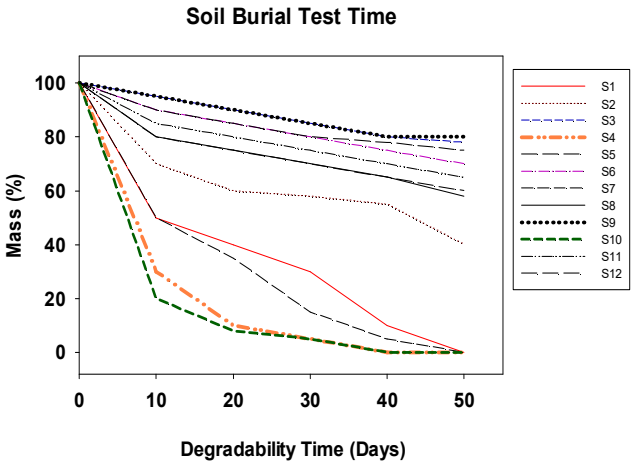

Fig. 5 Soil Burial Test Time

In detail, this study measured the degradation time of each sample from a total of twelve combinations, as presented in Fig. 5 above. The fastest time to degrade 100% completely was 40 days were obtained from samples S4 and S10, and the longest time to remove all wastes of biodegradable plastics were came from samples S3 and S9. Meanwhile, compared to this study, bioplastic from Sago starch done by the former group utilized gelatin reported that the longest time to degrade almost completely was in 16 days [21]. Moreover, few studies reported degradation rates up to 60% in certain period of time [12], [25], [27]. Hence, it is suggested that the results from this study are comparable and effective in removing the wastes created upon producing biodegradable plastic.

IV. Conclusion

The combination of glycerol, chitosan, and acetic acid affected the quality of bioplastic produced. The glycerol as a plasticizer increased the elongation of break and softened the structure of biodegradable plastic. The chitosan effect was seen by improving the tensile strength and making the product compact and rigid. Moreover, the presence of acetic acid tended to improve the mixing of slurry through a hydrolysis reaction. As the biodegradable plastic produced was easy and faster to degrade in nature, more studies need to be expanded to optimize the abundant feedstock in Papua.

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