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Production of bioplastics from organic waste with tapioca flour and glycerol

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Abstract. *Bioplastic is plastic that can be decomposed because it can return to nature. This study investigates the optimal composition for the production of bioplastics with various compositions of raw material. In this study, two types of organic waste (banana peel and rice waste), tapioca flour, and glycerol, were used in different proportions for the production of bioplastics. Comparison of the composition of banana peel:tapioca flour:glycerol, respectively 1:10:7.5 (sample A) and 1:13:11.25 (sample B). The ratio of the composition of rice waste:tapioca flour:glycerol in sample C and D is 1:10:7.5 and 1:13:11.25, respectively. Bioplastics are processed using a composite of banana peel or rice waste which is dried to a moisture content of 70%. 30 mL of distilled water was added to the bioplastic and heated until thickened. The bioplastic is molded in a baking sheet while still hot and in an oven at 117˚C, then cooled at room temperature. The optimal composition of bioplastic is found in sample B with a biodegradation test value of 58%, and it contains bioplastics with functional groups O-H, C-H, C=O, C=C, C-O, and =C-H in the FTIR test results. The quality standard values for the tensile strength test and elongation test at break in sample B have values of 10.9 MPa and 29%, respectively.*

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INTRODUCTION

Bioplastics are plastics that can be decomposed by the microorganisms' activity quickly (Kelibay, 2020). Bioplastics can decompose without leaving toxic residues after being used up because of their nature that can return to nature. Production of bioplastics with high starch and tensile strength is a requirement in the manufacture of bioplastics (Kelibay, 2020). Inggaweni and Suyatno (2015) stated that high starch accelerates bioplastic's biodegradation because starch is a food source for microorganisms.

Banana peel has a reasonably high starch value 59% (Melani *et al*., 2019). Widyaningsih *et al.* (2012) stated that banana peels contain 0.98% starch, 91.50% organic matter and affect tensile strength. Utami and Widiarti (2014) reported an addition of banana peel affected the tensile strength and elongation at break. Production of bioplastics using banana peel has been successfully investigated by Utami and Widiarti (2014), Munawaroh (2015), Agustin and Padmawijaya (2016), Melani *et al*. (2019).

Rice waste obtained from leftover rice that has been consumed and is not suitable for consumption also has a reasonably high starch. Bahari and Cahyonugroho (2018) reported bioplastic production using rice waste makes the plastic easy to decompose. The results of the tensile strength and elongation at the break test are higher, the biodegradation time is more optimal. Bioplastic using rice waste research has also been carried out by Aini *et al*. (2015), Marichelvam *et al*. (2019) and Harimbi and Satria (2020).

The use of starch in the production of bioplastics can improve its biodegradable properties. On the other hand, the complex and robust properties that occur in bioplastics are due to the amount of starch that is too much. The high amount of solids also create cross-links between starch polymers which are tightly formed so that even greater forces are needed to suppress the bioplastic process (Suryanto *et al.*, 2016). Tapioca flour is pure starch obtained by extraction from cassava milling, the amylopectin content will provide optimal stickiness (Azieyanti *et al.*, 2019). Kumoro and Purbasari (2014) have successfully carried out bioplastics made from tapioca flour and aking rice (Haryanto and Saputri (2016); Haryanto and Titani (2017); Karim and Musta (2019).

Starch as a primary material for bioplastics still has shortcomings, so additives are needed to improve its properties. A plasticizer is an additional material that serves to increase the elasticity properties. Research conducted by Suryanto *et al.* (2016) and Kelibay (2020) succeeded in the production of bioplastics addition glycerol. Kelibay (2020) states that glycerol is needed as a plasticizer that functions to increase the elasticity of bioplastics. Coniwanti *et al.* (2014) reported that glycerol could increase plastic films' flexibility and solubility, thereby increasing the biodegradability of starch-based plastic films. Suryanto *et al.* (2016) explained that glycerol has an essential role in manufacturing bioplastics because glycerol can reduce the hardness of bioplastics caused by too much tapioca flour content. Making bioplastics by utilizing waste banana peels and rice into more practical and environmentally friendly products supports the reduction of conventional plastic waste. This study aims to examine the optimal composition for the manufacture of bioplastics with various compositions of raw material for a banana peel or rice waste.

MATERIALS AND METHODS

This study was conducted at Solid Waste Laboratory, Department of Environmental Engineering, Engineering Faculty, Universitas PGRI Adi Buana Surabaya from February until August 2021.

Banana Peels and Rice Waste

Two types of organic waste for bioplastic production are banana peels and rice waste from household waste in Sidoarjo District, East Java Province, Indonesia. Eight experimental conditions were tested in duplicate using laboratory-scale reactors. The experimental design comprised two types of organic waste (banana peels and rice waste) and addition (tapioca flour and glycerol). Detailed experiment condition is given in Table 1. The experimental procedures were preparing banana peels and rice waste by mixing up with tapioca flour and glycerol. The banana peels and rice waste were air-dried for two days in the laboratory at room temperature before being used. The moisture content of banana peels and rice waste of 60-70%. Banana peels waste and rice waste are mashed and filtered through a sieve measuring 80 mesh particles.

Tabel 1 Experiment condition

Sample Code	Raw Material Composition (weight, gram)			
	Banana Peel Waste (BP)	Rice Waste (RW)	Tapioca Flour (TF)	Glycerol (G)
A (1 BP:10 TF:7.5 G)	1	-	10	7.50
B (1 BP:13 TF:11.25 G)	1	-	13	11.25
C (1 RW:10 TF:7.5 G)	-	1	10	7.50
D (1 RW:13 TF:11.25 G)	-	1	13	11.25

Production of Bioplastics

The production of bioplastics: 1 gram of banana peels and rice waste was placed in a beaker and weighted. Add tapioca flour (10 gram and 13 gram) and glycerol (7.5 gram and 11.25 gram) according to the variation. Add 30 mL of aqua dest in each variation of the composition, mix all the materials, and then put in *hotplate* *magnetic stirrer*. The boiling took about 20 minutes and was observed by using a stopwatch. The mixture then was stretched and pressed on over paper and was dried in the oven at a temperature of 120^{o}C. The mixture is then allowed to cool.

Bioplastics Analysis

The bioplastics analysis are tensile strength, elongation at break, biodegradability, and Fourier Transform Infra-Red Spectrophotometry (FTIR) according to ASTM or can be tested in laboratories that have implemented ISO/IEC 17025 (ASTM D882-12, 2002). The bioplastics product compared with bioplastics quality standards according to SNI 7188.7:2016 (Table 2). Tensile strength and elongation at break are physical reactions of bioplastics. The tensile test was done to carry out with the test object drawn from two directions so that the diameter shrinks and length increases. The amount of load and size increased during the test. Tensile strength is the maximum load accepted by the material ((Sofiah *et al.*, 2019). Percent lengths can be calculated by comparing the film's size at break and the film's size before being pulled by the elongation at break. Tensile strength and elongation at break can be calculated using equations (1) and (2), respectively.

$$\text{Tensil Strength (MPa)} = \frac{\text{Load of break}}{\text{(Original width)(original thickness)}} \tag{1}$$

$$\text{Elongation (\%)} = \frac{\text{(The final length of the test object} - \text{the initial length of the test object)}}{\text{The initial length of the test object}} \times 100\% \tag{2}$$

The biodegradation by utilizing microorganisms to determine biological reactions. Biodegradation test by preparing a 2 cm x 6 cm sample and weighing. The sample is buried for one week, then dried and weighed. The biodegradation test can be calculated using equation (3). FTIR analysis uses infrared wavelengths to determine chemical compounds or functional groups in bioplastics (Suryati *et al*., 2017).

$$\text{Weight residual (\%)} = \frac{\text{(The initial weight before biodegradation test} - \text{the final weight after biodegradation test)}}{\text{The initial weight before biodegradation test}} \times 100\% \tag{3}$$

Table 2 Bioplastics standard

No.	Parameter	Quality Standards*
1.	Tensile strength	Minimum of 13.7 Mpa (139.74 N/mm^2)
2.	Elongation at break	Maximum of 5%
3.	Biodegradation	Minimum 60%

* Indonesia National Standards SNI 7188.7:2016 about product category easy plastic and bioplastic shopping bags decompose

RESULTS AND DISCUSSION

Tensile Strength Value

The tensile strength of bioplastics in all treatments are shown in Figure 1 and Figure 2. Figure 1 shows that the addition of tapioca flour and glycerol give different tensile strengths in bioplastics. The tensile strength value using banana peel in samples A and B were 6.31 MPa and 10.9 MPa, respectively. In rice waste, samples C and D have tensile strength with a value was 5.57 MPa and 7.79 MPa, respectively. Tensile strength value tended that the sample using more addition tapioca flour and glycerol (13 gram and 11.25 gram) in samples B and D were higher than samples A and C using tapioca flour and glycerol (10 gram and 7.50 gram).

Inggaweni and Suyatno (2015) reported bioplastic production from composites High Density Polyethylene (HDPE) and cassava peel starch have tensile strength of 19.44 MPa. Wattimena *et al.* (2016) stated addition glycerol also effects on the tensile strength of bioplastics. Kumoro and Purbasari (2014) reported bioplastic production using addition glycerol 15% has a tensile strength value of 20.65 MPa. Tensile strength in all treatments did not meet the bioplastic quality standard (SNI 7188.7:2016), where the tensile strength value should be a minimum of 13.7 MPa. Sample B is closest to the minimum bioplastic quality standard.

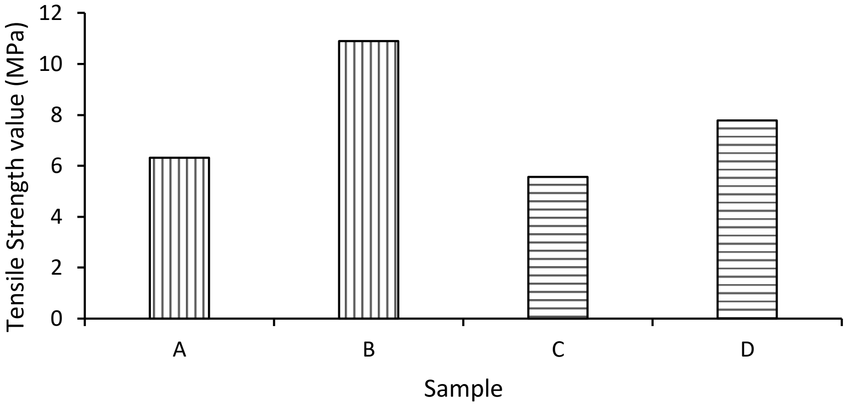

Figure 1 Tensile strength of bioplastic product

Elongation at Break Value

Elongation at a break in all treatments showed different values (Figure 2). Sample A and B using banana peel have elongation at break was 33% and 29%, respectively. Sample C and D using rice waste have an elongation at break were 33% and 60%, respectively Sinaga *et al.* (2014) reported that the more tapioca flour and glycerol affect elongation at break bioplastic. Inggaweni and Suyatno (2015) stated the elongation at break increased along with the increase in the amount of starch. The increased elongation at the break due to the addition of the amount of starch caused interaction between the two polymers (Layuk *et al.*, 2019). Elongation at a break in all treatments did not meet the bioplastic quality standard (SNI 7188.7:2016), where the elongation at break value should be maximum of 5%.

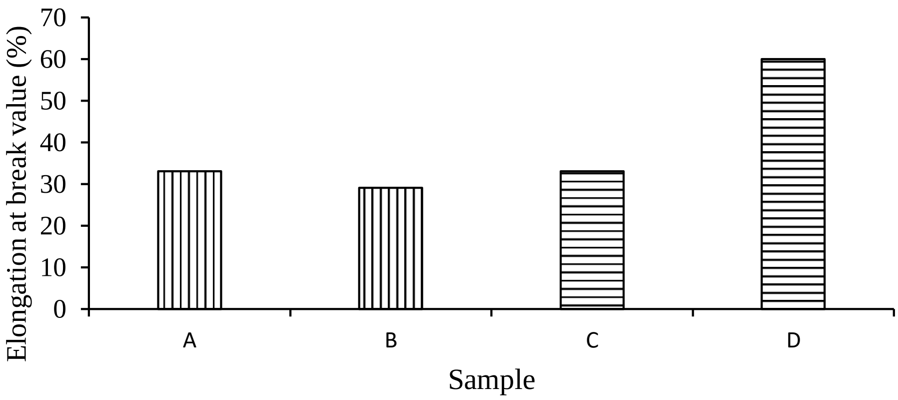

Figure 2 Elongation at break of bioplastic

Biodegradation Value

Biodegradation value in all treatments is shown in Figure 3. Figure 3 shows that biodegradation values in all treatments have varying values with a range of 58-87%. The biodegradation value from the banana peel in samples A and B were 67% and 87%, respectively. Samples C and D containing rice waste have 58% and 79% biodegradation values, respectively. Both biodegradation values in samples B and D using tapioca flour and glycerol were 13 grams and 11.25 grams have higher biodegradation values than samples A and C. It means that the ability to absorb water is more elevated and optimal microbial work process so that degraded quickly of bioplastic (Sofiah *et al.*, 2019). Haryanto and Saputri (2016) stated that the most essential property of bioplastics was decomposition.

Melani *et al.* (2019) reported biodegradation value of bioplastic from the banana peel with a range of 46.00-59.40% in 8 days. Haryanto and Saputri (2016) reported the highest biodegradation using soil media in 2 days with a value of 50%. Biodegradation values in samples A, B, and D met the bioplastic quality standard (SNI 7188.7:2016), where the biodegradation value should be minimum of 60%.

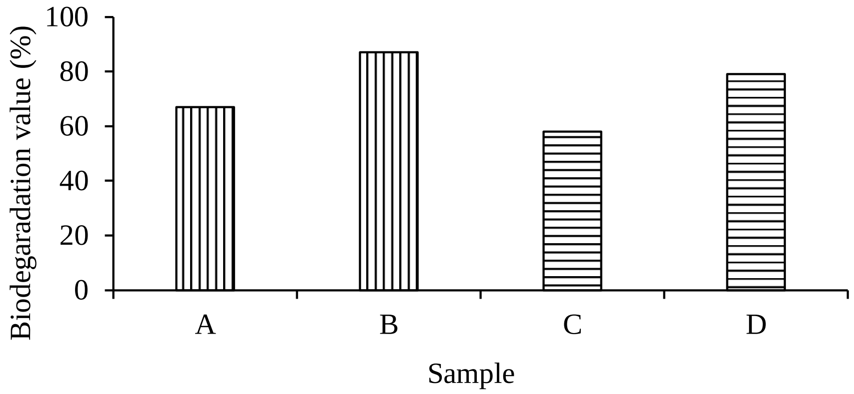

Figure 3 Biodegradation of bioplastic product

FTIR Value

FTIR value in all treatment shown obtained 11 kinds of compounds that had various peak values (Figure 4, 5, 6, and 7), containing *dextrose monohydrate powder*, where this compound is a white powder containing starch. *Hydroxypropyl-beta-cyclodextrin* is a compound that is difficult to dissolve in water. While the peak values for bioplastics in all treatments are about 400, which means that bioplastic products in all treatments are easily soluble in water because they have a higher value than the bioplastic quality standard. In addition to containing these two compounds, the bioplastic also contained *allyl alcohol*, *2-butene-1.4 diol*, *glucose*, *methyl-13c alcohol*, *propargyl alcohol*, *somaltose approx* 99%, *alpha-cyclodextrin hydrate*, *maltrotriose hydrate*, and *gamma-cyclodextrin hydrate*. The functional group O-H, C-H, C=O, C=C, C-O, and =C-H in all treatments indicate the formation of banana peel and rice waste bioplastics has already occurred, which was confirmed by FTIR spectroscopy. No indication of heavy metal, Cd<0.5 ppm, Pb<50 ppm, Hg<0.5 ppm, Cr^{6+}<50 ppm, and did not contain azo dyes.

Kumoro and Purbasari (2014) reported production of bioplastic from tapioca flour and rice flour resulted in FTIR showing the presence of an amide I group (a protein containing C=O bonds) and a spectral energy band with a position of cm^{-1} is a marker of the existence of amide III groups found at 1 200 and 1 350 cm^{-1}. Zaroh and Widyastuti (2019) reported bioplastic production from tapioca pulp to have FTIR results with functional O-H, C-H, C=O, C=C, C-O, and = C-H, it means that bioplastic can be graded because bioplastic material with 30% chitosan and temperature value of 95˚C.

Bioplastic Product from Banana Peels and Rice Waste

Production of bioplastics from banana peels and rice waste are shown in Figures 8, 9, 10, and 11. The bioplastic in samples A and B have a plastic thickness of ± 0,04 mm, brown in color, had was slightly sticky, easy to fold, and not easy to tear. The bioplastic in samples C and D have a plastic thickness of ± 0.07 mm, slightly cloudy white, has a very rubbery, sticky texture, is easy to fold, and is easily torn. Production of bioplastics from banana peels and rice waste can be alternative to existing conventional plastics, especially packaging applications.

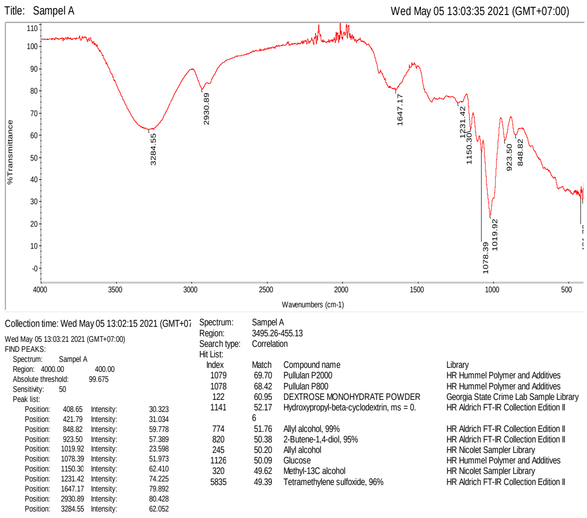

Figure 4 FTIR in sample A

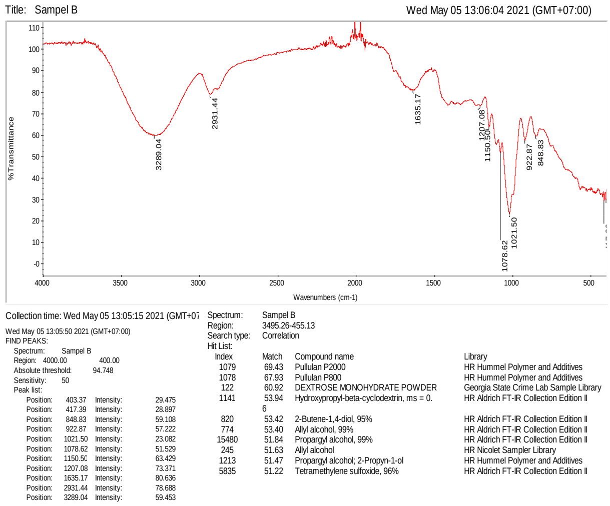

Figure 5 FTIR in sample B

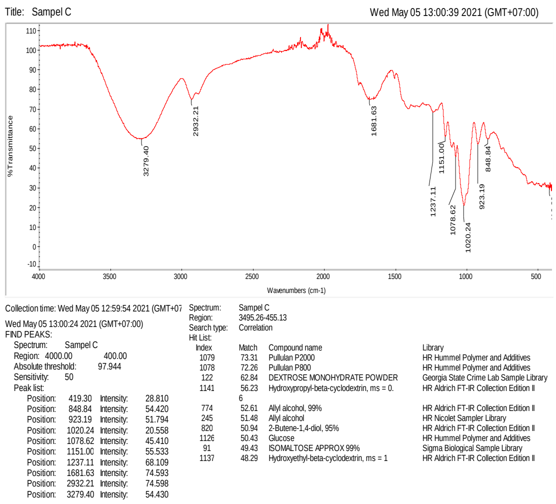

Figure 6 FTIR in sample C

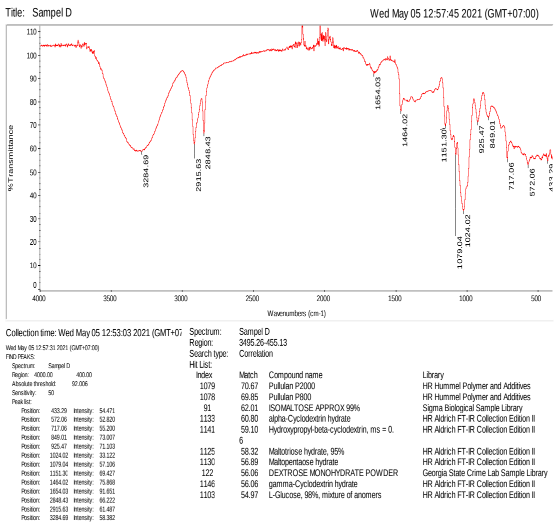

Figure 7 FTIR in sample D

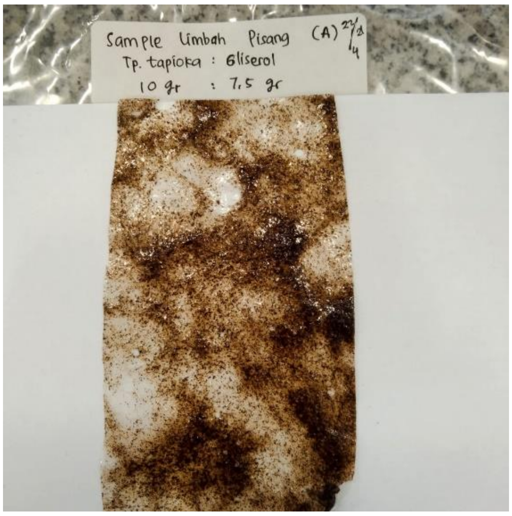

Figure 8 Bioplastic product in sample A

Figure 9 Bioplastic product in sample B.

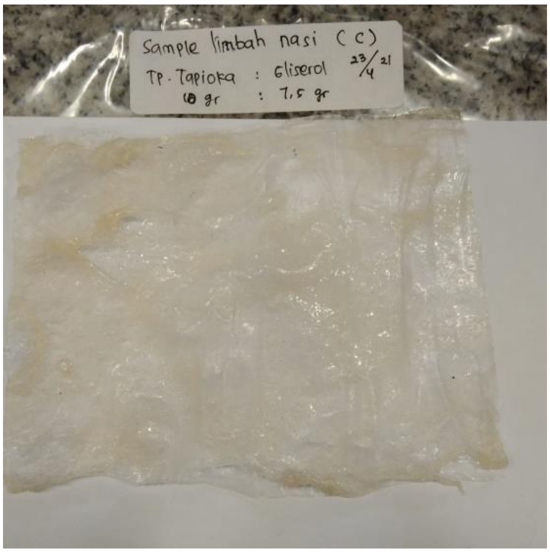

Figure 10 Bioplastic product in sample C

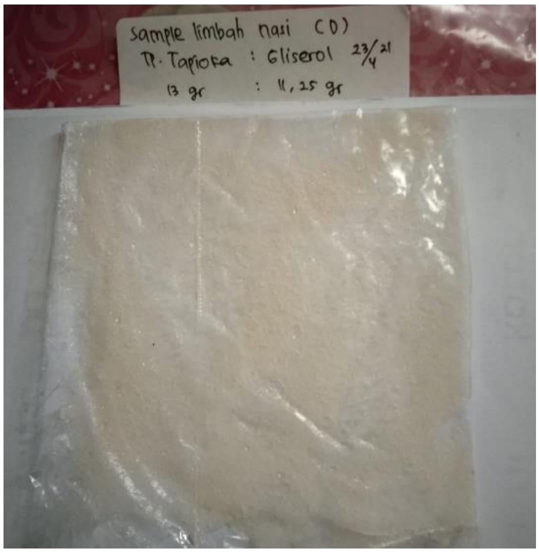

Figure 11 Bioplastic product in sample D

CONCLUSION

The optimum composition production of bioplastics occurred in sample B with banana peels:tapioca flour:glycerol was 1:13:11.25 (weight). Tensile strength and elongation at break of bioplastics were 10.9 MPa and 29%, respectively. The biodegradation value was 58%. Bioplastic content with functional group O-H, C-H, C=O, C=C, C-O, and =C-H on FTIR test. Biodegradation value met the requirement of bioplastics quality according to Indonesia National Standards No. 7188.7:2016. Both parameters of tensile strength and elongation at the break did not meet the requirement of bioplastics quality.

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