The Effect of Jerbung Shrimp Waste Flour (Fenneropenaeus Merguienis de Man) and Glycerol Ratio on Bioplastic Characteristics

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Abstract

This study was conducted to determine the ratio of shrimp waste flour and glycerol to the characteristics of bioplastics and determine the best ratio of shrimp waste flour and glycerol to the characteristics of bioplastics produced. The design carried out was a Completely Randomized Design (CRD) with 5 levels of treatment and three replications so that 15 experimental units were obtained. The parameters observed were tensile strength, elongation, thickness, water resistance, water vapor transmission rate (WVTR) and degradation. The data obtained were analyzed using ANOVA at 5% and 1% levels. If there is a significant effect on the treatment, the Duncan's Multiple Range Test (DNMRT) is continued at the 5% level. The treatment given is the ratio of shrimp waste flour and glycerol with different ratios, namely P1 shrimp waste flour 0 g : glycerol 3 g ; P2 shrimp waste flour 0.5 g : glycerol 2.5 g ; P3 shrimp waste flour 1 g : glycerol 2 g ; P4 shrimp waste flour 1.5 g ; glycerol 1.5 g ; P5 shrimp waste flour 2 g : glycerol 1 g. The results showed that P3 with a ratio of 1 g: 2 g glycerol was the best treatment with a tensile strength value of 14.45 N/mm2, elongation of 84.09%, thickness of 0.35 mm, water resistance of 69.21%, wvtr of 20.80 g/m2.h and bioplastics decompose faster than synthetic plastic types.

Keywords : shrimp waste flour, glycerol and bioplastic

1. Introduction

One of the many wastes is plastic waste. Plastic waste is a global environmental problem today. Plastic is widely used in everyday life, because it has advantages such as strong, lightweight, economical and stable. However, the plastic circulating in the market today is one type of synthetic polymer made from petroleum which is difficult to be decomposed by nature. As a result, if more and more people use plastic, environmental pollution such as soil pollution will also increase. Therefore, it requires a solution to overcome this environmental problem, one of which is to develop biodegradable plastic materials (bioplastics) which means that plastics can be decomposed by microorganisms naturally into environmentally friendly compounds.[3]

Shrimp waste comes from the head, skin and tail of shrimp which contains good nutrition, namely 53.47% protein, 6.65% fat, 17.28% water, 7.72% ash and 14.61% chitin, so shrimp waste has the potential to be processed into shrimp waste flour which can be further utilized into biomaterials for making bioplastics.[10] Flour derived from shrimp waste goes through a deproteination and demineralization process to obtain chitin.[18] The next step that must be done is the deacetylation process to become chitosan.[17]

This chitosan material is hard and rigid so that plasticizers such as glycerol are needed to make it more flexible, in addition to obtaining plastic mechanical properties in the form of transparent, elastic, hydrophilic.[20] Plasticizer is a low molecular weight organic material added to the product, the purpose of which is to reduce the rigidity of the polymer and increase the flexibility and extensibility of the polymer.

The objectives of this research are to determine the effect of jerbung shrimp waste flour and glycerol on the manufacture of bioplastics and to determine the effect of the ratio of jerbung shrimp waste flour and glycerol that produces bioplastics with the best characteristics.

2. Research Methods

2.1 Tools and Materials

The materials used in this study were shrimp waste, purple uwi (Dioscorea alata), plasticizer (glycerol), distilled water, CH3COOH, and NaOH.

The tools used in the research are scales, dropper pipettes, beakers, measuring cups, volumetric flasks, magnetic stirrers, hot plates, flexi glass molds, blenders, spoons, sieves, porcelain cups, stirring rods, basins, ovens, verniers, pH meters. The tools used in testing are test tubes, Petri dishes, RH meters, desiccators, texture analyzers, and screw micrometers.

2.2 Bioplastic Formulation

The process of making bioplastics is carried out according to the modified method where the bioplastics in this study were made with 2 layers. bioplastic formulations can be seen in the following table.

Treatment	Shrimp Waste Flour (g)	Purple Yam Starch (g)	Glycerol (g)	Ch3COOH (ml)	Distilled Water (g)	Total (g)
1	0	5	3	5	137	150
2	0,5	5	2,5	5	137	150
3	1	5	2	5	137	150
4	1,5	5	1,5	5	137	150
5	2	5	1	5	137	150

2.3 Research Implementation

2.3.1 Preparation of starch from purple yam

The outer skin of the uwi tuber is cleaned and then washed with the aim of removing sap and remaining soil on the tuber, then cut and sliced with a thickness level of 2 to 3 mm, then washed again to remove mucus and then drained. The sliced uwi was soaked for 30 minutes with 15% salt water solution, then washed again with clean water 3 times. Next, the uwi is blended until smooth, the blended uwi is then filtered using a 200 mesh sieve. Then the suspension obtained was precipitated for 6 hours and then stored in a refrigerator at 4°C. Furthermore, the sediment obtained was rinsed using distilled water with the aim of purifying the starch. Then the starch was put into the oven to dry for 6 hours at 50°C. After the starch was dried, it was pulverized and sieved using a 60 mesh sieve and then stored in an airtight container.

2.3.2 Preparation of Shrimp Waste Flour

The manufacture of flour uses basic ingredients in the form of waste (skin, legs and tails) of shrimp species in good condition with the characteristics of having a hard texture, not emitting foul odor, still fresh, then shrimp waste is washed using running water with the aim of getting clean shrimp waste (no impurities). The cleaned shrimp waste was then homogenized and dried in an oven at 100°C for 5 hours. After the drying process is complete, then pulverization is carried out using a blender and then sieved with a 60 mesh sieve. Next, the shrimp waste flour was weighed as much as 90 grams and soaked using 10% CH_3COOH solution for 1 hour, then washed until the resulting pH was neutral, after which it was soaked with 0.5 M NaOH while stirring for 2 hours on a hot plate at 65°C. After the mixture cools, it is filtered and washed with distilled water until the pH is neutral. After the pH is neutral, it is dried in an oven at 100 ° C for 4 hours, then after drying is complete, sieving is carried out with a 60 mesh sieve.

2.3.3 Film Preparation

Making bioplastics is done by making the solution first, namely mixing 5% CH3COOH as much as 5 ml with shrimp waste flour based on treatment, soaked for 1 hour. Then aquadest and starch were added to the beaker glass, after which glycerol was added at the 10th minute. The solution was homogenized using a magnetic stirrer at 80 ℃ until the mixture became semi-solid. After obtaining the bioplastic solution, the film mixture was poured into a flexi glass mold measuring 20 cm x 20 cm. Then put in the oven, oven for 24 hours at 50 ℃, after which the mold is removed from the oven and cooled at room temperature. Then the finished film was peeled off and stored in an airtight container. Subsequently, the same process was carried out twice. After that, the two bioplastics were glued together using ± 15 g of bioplastic dough, then the bioplastics were baked for ± 5 hours at 50 ℃, after which the mold was removed from the oven and cooled at room temperature.

2.4 Characterization of Bioplastic

2.4.1 Tensile Strength

The tensile strength was measured using the Texture Analyzer. The tensile strength is determined based on the maximum load at which the film breaks. The equation for tensile strength is as follows:

Tensile Strength = $\frac{Force\ (F)}{In\ the\ width}\ x\ 15$

2.4.2 Elongation

Elongation or percentage elongation is measured using the Texture Analyzer. Elongation or percentage elongation is based on the elongation of the film when the film breaks. The equation for percentage elongation is as follows:

% Elongation = $\frac{(A-B)}{B} x\ 100\%$

Description: A = Length after breaking (mm)

B = Length before breaking (mm)

2.4.3 Thickness

The film thickness was measured using a screw micrometer with an accuracy of 0.001 mm. The film thickness was measured five times at different position points and the average value was taken.

2.4.4 Water Vapour Transmission Rate (WVTR)

The test tube containing 5 grams of calcium chloride (CaCl) was covered using a 6x6 cm film, then folded at the mouth of the test tube until it was tightly closed and the weight of the test tube was weighed. The tube was placed in a desiccator that was saturated with saturated sodium chloride (NaCl) (75% RH). Every 30 minutes the test tube was weighed again, the change in the tube was then recorded and plotted as a function of time. The calculation of WVTR can use the following formula:

WVTR = $\frac{Slope}{A}$

Description:
WVTR = Water Vapor Transmission Rate Analysis g/m^2.hour
Slope = Linear function of weight gain and time (g/hour)
A = Film area (m^2)

2.4.5 Water Absorption

The initial weight was weighed (W_0). Then the sample was dipped in distilled water for 20 minutes, then weighed again to get the final weight (W). Before calculating the water resistance value, first calculate the absorption experienced by bioplastics.

The absorption experienced by the film is then calculated using the following formula:

Water absorption (%) = $\frac{W-W0}{W0} x\ 100\%$

Description:
W_0 : Initial weight of the sample
W : Weight of the sample after immersion

2.4.6 Degradation

Bioplastic degradation testing is by burying bioplastics in the soil, each sample is cut with a size of 2 x 2 cm. Observed the degradation rate of burial in the soil for 7 days by checking on the 1st, 3rd, 5th and 7th days.

2.4.7 Data Analys

The data obtained were analyzed using Analysis of Variance (ANOVA) at the 5% and 1% levels. If there are significantly different data results, then continued with further tests, namely the Duncan New Multiple Range Test (DNMRT) test. And biodegradation data were analyzed descriptively by displaying the results during testing in the form of images.

3. Results and Discussion

3.1 Product Description

Bioplastics are biodegradable plastics or environmentally friendly plastics that can be decomposed by the activity of microorganisms. This bioplastic is partly or almost all of its components come from renewable raw materials.[8.1] The advantage of using bioplastics as food packaging is that it can extend product shelf life and is environmentally friendly.[14] Bioplastics are not intended to completely replace synthetic packaging, but they have the potential to reduce packaging and limit the transfer of water and aromas between packaged food components.

Based on the research that has been done, bioplastics with the ratio of shrimp waste flour and glycerol in this study have a tensile strength value that is still classified as good to withstand the maximum load, elongation or percentage of elongation which increases with the addition of glycerol and decreases the amount of shrimp waste flour, appropriate thickness, water resistance increases with the addition of the amount of shrimp waste flour and decreases the amount of glycerol so that bioplastics are resistant to water and not easily destroyed or damaged, the value of the water vapor transmission rate is getting lower and bioplastics are quickly decomposed in the soil compared to synthetic plastics. The following is an image of bioplastic products with a ratio of shrimp waste flour and glycerol:

Flour : Glycerol

(0 g : 3 g)

Flour : Glycerol

(0,5 g : 2,5 g)

Flour : Glycerol

(1 g : 2 g)

Flour : Glycerol

(1,5 g : 1,5 g)

Flour : Glycerol

(2 g : 1 g)

3.2 Tensile Strength

Tensile strength is the maximum pull that can be achieved until the bioplastic remains before breaking. Tensile strength is a mechanical property of bioplastics and knows the amount of force required to achieve maximum pull on each area of the bioplastic film or to determine the strength of the film. The greater the tensile strength, the better the film is in resisting mechanical damage. The average value of the tensile strength of the bioplastic ratio of shrimp waste flour and glycerol can be seen in the table below.

Shrimp Waste Flour : Glycerol (g)	**Tensile Strength (N/mm^2) ± SD**
0 : 3	$5,97 \pm 0,85^{a}$
0,5 : 2,5	$11,81 \pm 2,06^{ab}$
1 : 2	$14,45 \pm 2,60^{bc}$
1,5 : 1,5	$20,91 \pm 7,53^{c}$
2 : 1	$21,74 \pm 5,15^{c}$

Notes : The numbers followed by the same lowercase letters are not significantly different at the level of 5% level according to the DNMRT test.

Based on the results of analysis of variance, the ratio of shrimp waste flour and glycerol has a very significant effect on the average value of tensile strength of bioplastics. The average value of bioplastic tensile strength ranged from 5.97 N/mm^2 – 21.74 N/mm^2. Table 3. Shows that the lowest average value of tensile strength is found in the treatment of shrimp waste flour and glycerol ratio (0 g: 3 g) which is 5.97 N/mm^2 and the highest value is found in the treatment of shrimp waste flour and glycerol ratio (2 g: 1 g) which is 21.74 N/mm^2.

The mechanical properties of bioplastics are influenced by the amount of content of the constituent components of the film, namely shrimp waste flour and glycerol. The average tensile strength value of bioplastics increases as the mass composition of shrimp waste flour increases and the mass composition of glycerol decreases, and vice versa. The two-layer bioplastic also increased the tensile strength value of bioplastics, where bioplastics with one layer had a lower value. The higher the mass composition of shrimp waste flour, the more hydrogen bonds there will be in the film so that the chemical bonds of the film will be stronger and more difficult to break because it requires a lot of energy to break the bonds, this is caused by bioplastic particles undergoing many physical changes so that the film is more homogeneous and the structure is tight, with these characteristics making the tensile strength value even greater.[8] Tensile strength is also influenced by the plasticizers added in the filmmaking process.[19] Tensile strength and plasticizer efficiency depend on the molecular weight. Glycerol has a low molecular weight of 92.09 so it is easy to enter the polymer chains of proteins and polysaccharides, increasing the flexibility of the film. And plasticizers can reduce the intermolecular force of polar groups (-OH) around the plasticizer chain is believed to develop plastic polymer hydrogen bonds that replace polymer-polymer interactions in biopolymer films. Increasing the amount of glycerol will result in a reduction of intermolecular interactions and an increase in the movement of the polymer chains, so the tensile strength will decrease.[7]

The results in this study are in accordance with JIS 2019 class 5 standard, where bioplastics have a tensile strength value below 25 N/mm2. This result is higher than the tensile strength produced by the research of Albar, et al., (2021) made from glycerol and chitosan 0 – 3% is 1.5369 Mpa.

3.3 Elongation

Plasticizers are low molecular weight substances that can enter into the polymer matrix of proteins and polysaccharides thereby increasing film flexibility and film-forming ability. Tensile tests on bioplastics cause changes in the length of the sample called elongation. Glycerol as a plasticizer can give elastic properties to plastics. Films made from starch alone are more elastic and have low tensile strength and percent elongation.[9] The average value of elongation or percent elongation of the ratio of shrimp waste flour and glycerol can be seen from the table below.

Shrimp Waste Flour : Glycerol (g)	**Elongation (%) ± SD**
0 : 3	109,77 ± 3,90^{b}
0,5 : 2,5	85,28 ± 24,95^{ab}
1 : 2	84,09 ± 26,20^{ab}
1,5 : 1,5	67,90 ± 2,43^{a}
2 : 1	62,43 ± 0,30^{a}

Notes : The numbers followed by the same lowercase letters are not significantly different at the level of 5% level according to the DNMRT test.

Based on the results of the analysis of variance, it shows that the ratio of shrimp waste flour and glycerol has a very significant effect at the level of $p < 0.01$. The average value of bioplastic elongation ranged from 62.43% - 109.77%. The elongation value of bioplastics shows the opposite of the tensile strength value of bioplastics. From table, it shows that the lowest average value of elongation is found in the treatment of shrimp waste flour and glycerol ratio (2 g: 1 g) which is 62.43% and the highest value in the treatment of shrimp waste flour and glycerol ratio (0 g: 3 g) which is 109.77%.

Percent elongation of plastic is inversely proportional to its tensile strength. Plastics that have the most C = C groups, the bond is difficult to release, so it tends to be difficult to stretch and have a smaller elongation value. This is due to the decreasing bond distance between the molecules.[14] Therefore, with the increase in the amount of shrimp waste flour and the reduction of glycerol, the elongation value decreases, and vice versa. The results of this study, the elongation value based on JIS 2019 is included in class 4, where in class 4 the elongation value ranges from 20% or more to reach 200%.

3.4 Thickness

Film thickness is one of the important parameters in bioplastics, because it can affect other parameters, such as water vapor transmission rate, tensile strength and water resistance. Thickness testing was measured using a 0.001 mm accuracy screw micrometer, taken from the average at 5 different points of the film. The average thickness of bioplastics with the ratio of shrimp waste meal and glycerol can be seen in the following table.

Shrimp Waste Flour : Glycerol (g)	**Thickness (mm) ± SD**
0 : 3	0,31 ± 0,04[a]
0,5 : 2,5	0,32 ± 0,04[a]
1 : 2	0,35 ± 0,03[ab]
1,5 : 1,5	0,39 ± 0,05[bc]
2 : 1	0,44 ± 0,07[c]

Notes : The numbers followed by the same lowercase letters are not significantly different at the level of 5% level according to the DNMRT test.

Based on the table above, the highest bioplastic thickness value is bioplastic with the treatment of shrimp waste flour and glycerol ratio (2 g: 1 g) which is 0.44 mm while the lowest value is in the treatment of shrimp waste flour and glycerol ratio (0 g: 3 g) which is 0.31 mm. These results are thicker than the standard results of the best thickness made from teak leaf extaract at 20% glycerol concentration, which is 0.18 mm.[19]

Based on the analysis of variance, it shows that bioplastics with the ratio of shrimp waste flour and glycerol are a very significant effect ($p<0.01$) on thickness. This shows that the higher the ratio of adding the amount of shrimp waste flour and reducing the amount of glycerol will increase the total solids in the solution. The increase in total solids in the solution causes the film thickness to increase.[15] This is in accordance with the opinion of Nugroho, et al. (2013) which states that an increase in the amount of solids in the solution results in more polymers that make up the film matrix.[16]

3.5 Water Absorption

Water resistance is done to determine the level of resistance of a bioplastic to water. This test is carried out by placing the bioplastic sample into a container filled with water then the sample is lifted and weighed until a constant weight of the bioplastic is obtained and the water absorption value is obtained. The greater the water absorption value, the lower the level of resistance of the bioplastic and the bioplastic is quickly damaged. Conversely, the lower the water absorption value, the higher the level of bioplastic resistance, which means that it is able to protect the packaged product and is not easily damaged or destroyed in water. The value of water resistance can be seen in the following table.

Shrimp Waste Flour : Glycerol (g)	**Water Absorption (%) ± SD**
0 : 3	63,71 ± 8,82
0,5 : 2,5	64,74 ± 3,35
1 : 2	69,21 ± 4,58
1,5 : 1,5	70,98 ± 2,60
2 : 1	79,37 ± 9,84

Water resistance is an important parameter of the nature of bioplastics, the value of water resistance is obtained by finding the value of water absorption first. The value of water resistance obtained is different. Based on the analysis of variance, bioplastics with the ratio of shrimp waste flour and glycerol have no significant effect on water resistance. It can be seen in the table above. With the addition of the amount of shrimp waste flour and reduced glycerol, the value of water resistance increases. The highest water resistance value in the bioplastic treatment with the ratio of shrimp waste flour and glycerol (2 g: 1 g) is 79.37% and for the lowest water resistance value in the bioplastic treatment with the ratio of shrimp waste flour and glycerol (0 g: 3 g) is 63.71%.

In this study, bioplastics with a mixture of starch and glycerol have a low water resistance value so that the solubility of bioplastics in water is fast which results in bioplastics being easily destroyed, this is because bioplastics are not resistant to water because starch and glycerol are hydrophilic so they tend not to be resistant to water. Glycerol as a plasticizer increases the flexibility of plastic, but the amount of empty space that increases also along with the increase in glycerol increases the gap to be occupied by water molecules, therefore the value of water absorption increases with increasing amounts of glycerol added.[13] However, the addition of shrimp waste flour in the manufacture of bioplastics increases the value of water resistance due to the content in shrimp waste flour which is hydrophobic. The greater the amount of addition of shrimp waste flour, the greater or increased water resistance value which means that this bioplastic is getting better against water.

3.6 *Water Vapour Transmission Rate* (WVTR)

Water Vapour Transmission Rate (WVTR) is the most important parameter in assessing film quality. Water Vapour Transmission Rate indicates the speed of penetrating water vapor (per gram per second) or the ability of the film to inhibit water vapor transmission, thus affecting the structure of water in the film which makes the physical properties decrease. The value of Water Vapour Transmission Rate (WVTR) or water vapor transmission of bioplastics with the ratio of shrimp waste flour and glycerol can be seen in the following table.

The water vapor transmission rate can be used to determine the shelf life of the product. If the water vapor transmission rate can be restrained, the shelf life of the product can be extended. Water loss in fruits and vegetables is a major cause of deterioration during storage. Water loss can cause fruits and vegetables to experience weight loss and appear wilted, making them less desirable to consumers.[22] Water vapor migration generally occurs in the hydrophilic part of the film. Thus, the ratio between the hydrophilic and hydrophobic parts of the film component will affect the water vapour transmission rate value of the film.[5]

The higher the WVTR value, the higher the permeability of the packaging, the more water vapor will escape from or enter the packaging.[6] A good film is one that is not easily passed by water vapor or has a low water vapor transmission rate value. The value of Water Vapour Transmission Rate (WVTR) or water vapor transmission of bioplastics with a ratio of shrimp waste flour and glycerol can be seen in the following table.

Shrimp Waste Flour : Glycerol (g)	**WVTR (g/m².Jam) ± SD**
0 : 3	28,66 ± 2,12
0,5 : 2,5	26,97 ± 2,18
1 : 2	20,80 ± 0,92
1,5 : 1,5	19,11 ± 1,49
2 : 1	16,18 ± 3,61

Based on the table above, the water vapor transmission rate of bioplastics shows that the highest water vapor transmission value in the treatment of shrimp waste flour and glycerol ratio (0 g: 3 g) is 28.66 g/m².hour while the lowest value in the treatment of shrimp waste flour and glycerol ratio (2 g: 1 g) is 16.18 g/m².hour. And it can be seen in the table above, that the water vapor transmission of bioplastics has decreased with the increase in the amount of shrimp waste flour and the reduction in the amount of glycerol.

This is because the plasticizer is hydrophilic and is able to reduce the intermolecular tension in the film matrix which causes the space between molecules to get bigger so that water vapor can penetrate the film. In addition, glycerol is a type of plasticizer that is hydrophilic, adds polar properties and is easily soluble in water, so that with a low amount of glycerol, it reduces the water vapor permeability of the resulting film. The addition of plasticizers such as glycerol will also cause a decrease in internal hydrogen bonds and an increase in intermolecular distances which leads to an increase in film permeability.[12] In addition, decreased intermolecular interactions and increased molecular mobility will facilitate the migration of water vapor molecules.[23] The inhibition characteristics of the film are influenced by its hydrophilic or hydrophobic nature as well as the grade and suitability of the plasticizer used.

3.7 Degradation

Degradation testing is carried out to determine the level of resistance of a bioplastic to mold growth, or to determine the length of time the bioplastic decomposes. Plastics made from natural (organic) materials tend to have relatively short durability and shelf life.[27] In this study, the degradation process of bioplastics in each bioplastic is different.

Day	Bioplastic				
	Shrimp Waste Flour (g) : Glycerol (g)				
	(0 : 3)	(0,5 : 2,5)	(1 : 2)	(1,5 : 1,5)	(2 : 1)
16					
24					

The ratio of jerbung shrimp waste flour and glycerol (0 g: 3 g) degraded faster than the other ratios of jerbung shrimp waste flour and glycerol. Glycerol plasticizer is hydrophilic, so the sample with the most glycerol takes faster time to degrade. In the ratio of shrimp waste flour and glycerol (2 g: 1 g), having the highest amount of shrimp waste flour addition, it takes longer to degrade. This is because it has the highest intensity of the C = C group, which causes the intermolecular bonds of the plastic to be stronger and more difficult to break. In addition, the content contained in jerbung shrimp waste flour is hydrophobic, so bioplastics with the highest amount of jerbung shrimp waste flour addition take longer to degrade in the environment.[11]

4. Conclusions and Discussion

4.1 Conclusions

Based on the results of the research that has been carried out, it can be concluded that the ratio of shrimp waste flour and glycerol to the manufacture of bioplastics has a significant effect on the tensile strength test, elongation or percent elongation and thickness, but has no significant effect on the value of water resistance, water vapour transmission rate (wvtr) or water transmission rate and does not take a long time to degrade and the best bioplastic is found in the ratio of shrimp waste flour and glycerol (1 g: 2 g) with a tensile strength value of 14.45 N/mm^2, elongation 84.09%, thickness 0.35 mm, water resistance 69.21%, wvtr 20.80g/m^2.h and this bioplastic is faster to decompose into the environment than synthetic plastic types.

4.2 Discussion

From the results of the study it is suggested for further research that a transparency test should be carried out on bioplastics in order to determine the transparency of the film to be accepted by consumers of packaged products and continued with the application of bioplastics to food ingredients such as pempek.

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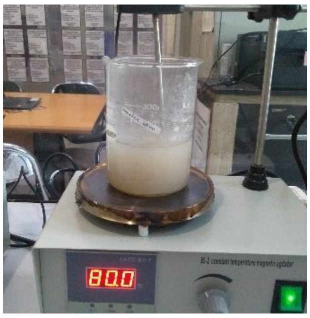
Documentation

		
Purple Yam	Purple yam starch	Shrimp waste
		
Flour sieving	Shrimp waste flour	Heating to 80°C
		
Oven	Removal of film from mold	Bioplastic