THE USE SKIN STARCH OF JACKFRUIT (*Artocarpus heterophyllus*) AND CHITOSAN IN A BIOPLASTIC PRODUCTION WITH THE ADDITION OF VIRGIN COCONUT OIL (VCO) AS PLASTICIZER

Pemanfaatan Pati Kulit Nangka dan Kitosan pada Produksi Bioplastik dengan Penambahan Virgin Coconut Oil (VCO) sebagai Plasticizer

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Abstrak: *Penelitian ini bertujuan menentukan konsentrasi terbaik pada perbandingan pati kulit nangka dan kitosan dengan penambahan virgin coconut oil (VCO) sebagai plasticizer serta karakteristik bioplastik. Metode penelitian terdiri dari tahapan sebagai berikut: ekstraksi pati kulit nangka yang menggunakan dua proses yaitu proses delignifikasi dan proses bleaching. Pembuatan bioplastik dengan perlakuan sebagai berikut: Pati kulit nangka: kitosan masing-masing dengan perbandingan P1 = 100%:0%; P2 = 85%:15%; P3 = 70%:30% dan P4 = 55%:45% dengan penambahan VCO sebesar 10% dari total pati kulit nangka dan kitosan dan dilakukan pengadukan selama 1 jam hingga homogen. Larutan yang dihasilkan kemudian dicetak pada cetakan kaca dengan ukuran 20 x 15 cm dan dikeringkan dalam oven pada suhu 60 ^{o}C selama 6 jam. Bioplastik didinginkan pada suhu kamar selama 6 jam kemudian dilepaskan dari pelat kaca dan distabilkan dalam desikator sebelum dianalisis. Analisis data menggunakan metode Rancangan Acak Lengkap (RAL) dengan 4 taraf perlakuan dan tiga kali ulangan. Hasil penelitian diperoleh bahwa penambahan kitosan dan VCO berpengaruh sangat nyata ($p<0.05$) terhadap karakteristik bioplastik. Rasio perlakuan terbaik yang diperoleh pada perbandingan pati kulit nangka : kitosan 55%:45% dengan nilai ketebalan 0,1682 mm, kuat tarik 0,5933 MPA, daya serap air 20% dan uji biodegrabilitas sebesar 50% yang dapat terdegradasi selama 3 minggu.*

Kata Kunci: *Bioplastik, pati kulit nangka, kitosan, virgin coconut oil*

Abstract: *This study aims to determine the best concentration on the comparison of jackfruit skin starch and chitosan with the addition of virgin coconut oil (VCO) as a plasticizer and the characteristics of bioplastics. The research method consisted of the following steps: extracting jackfruit skin starch using two processes, namely the delignification process and the bleaching process. The producing of bioplastics was carried out with the following treatments: Jackfruit skin starch: chitosan, each with a ratio of P1= 100%:0%; P2 = 85%:15%; P3 = 70%:30% and P4 = 55%:45% with the addition of VCO of 10% of the total starch of jackfruit skin and chitosan and stirred for 1 hour until homogeneous. The resulting solution was then printed on a glass mold with a size of 20 x 15 cm and dried in an oven at 60^{o}C for 6 hours. The bioplastics were cooled at room temperature for 6 hours then removed from the glass plate and stabilized in a desiccator before being analyzed. Data analysis used Completely Randomized Design (CRD) method with 4 treatment levels and three replications. The results showed that the addition of chitosan and VCO had a very significant effect ($p<0.05$) on the characteristics of bioplastics. The best treatment ratio was obtained in the ratio of jackfruit skin starch: chitosan 55%:45% with each thickness value of 0.1682 mm, tensile strength of 0.5933 MPA, 20% water absorption and 50% biodegradability test which can be degraded for 3 weeks.*

Keywords: *Bioplastic, jackfruit skin starch, chitosan, virgin coconut oil*

INTRODUCTION

The increase in the volume of plastic waste in Indonesia is a very serious problem that has not been resolved until now. Meanwhile, with the increase in population, the volume of plastic waste generated from human activities (Kumar R et al., 2021). Indonesia is ranked second in the world after China in producing plastic waste in the waters, which reachs 187.2 million tons. The problem with plastic waste is that if the number is increasing in the environment, it will contribute to the environmental pollution, especially the soil. Given that the nature of plastic is difficult to decompose in the soil, the plastic will decompose in more than 20 years and can even reach 100 years so that it can reduce the level of soil fertility (Alabi A Okunola et al., 2019).

Bioplastic is an innovation that was created to help reduce the problem of plastic waste by creating a plastic that can be easily decomposed. Thus it does not cause waste buildup. Bioplastics are typically made from plant parts containing starch and cellulose. Starch is used as the primary ingredient in the production of bioplastics because it contains amylose and amylopectin, which can produce bioplastics with good physical properties that decompose easily when released into the environment (Nor Izaida Ibrahim et al., 2021).

In Indonesia, jackfruit is a tropical fruit that has no seasons and can grow in almost part of the country (Anna et al, 2017). Jackfruit is used as a vegetable and as a food additive in Indonesia (Hamidah et al, 2015). There are various kinds of jackfruit processed products include chips, jackfruit lunkhead, fruit juice, pasta, and sweets. The flesh of the fruit is only used in the processing; the rest is in the form of fruit skin waste and jackfruit straw (Akter F et al., 2019).

The cellulose content of jackfruit is 38.69% (Wulandari et al., 2015) and Hermawani et al., 2019). Jackfruit skin carbohydrates consist of glucose, fructose, sucrose, starch, fiber and pectin with a total amount of 15.87%. Jackfruit skin protein is 1.30% (Ibna Suli T R et al., 2021). Jackfruit straw contains 76.24% water, 0.53% ash, 1.0% protein, 0.60% fat and 15.87% carbohydrates (Ibna Suli T R., 2021). Jackfruit rind and straw with carbohydrate content in the form of sugar and cellulose can be used as flour (Harimbi S et al, 2020).

To improve the quality of bioplastics, elastic additives such as plasticizers are commonly used. Plasticizers such as glycerol and sorbitol are commonly used in the production of bioplastics because they are inexpensive and easy to obtain. Glycerol is frequently used as a plasticizer because it has advantages such as increasing the flexibility and elasticity of bioplastics and providing flexibility to the starch structure so that it can be shaped (Kumoro and Purbasari, 2014). The addition of virgin coconut oil as a plasticizer in the producing of bioplastic from glutinous corn starch (Zea May) and chitosan resulted in a good tensile stress value of 0.5171 - 18, 1667 MPa. (Nur Safitri et al., 2021).

Production of bioplastics by utilizing skin waste from biomass is an alternative developed at this time such as the use of jackfruit skin. This research aims to determine the optimum concentration on the comparison of jackfruit skin starch with the addition of virgin coconut oil as a plasticizer and bioplastic characteristics.

METHODS

This research was conducted in January-March 2022 at the Laboratory of Analytical Chemistry, Faculty of Engineering, Bosowa University, Makassar.

The tool used for producing bioplastics from jackfruit skin starch and chitosan with addition of virgin coconut oil (VCO) as platisticizer are bowl, knife, blender, scissors, hot plate, analytical balance, stirer rod, erlenmeyer, tray, mesh siever, beaker, measuring cup, flannelette, flask, oven, plastic container, petri dish, spoon, dropper, screw micrometer, knife. The ingredients used in are jackfruit skin, chitosan, VCO, NaOH, hydrogen peroxide, acetic acid, aquades.

Experimental design

The research treatment is based on the preliminary study, as follows:

P1 = Jackfruit starch 100%: Chitosan 0%
P2 = Jackfruit starch 85%: Chitosan 15%
P3 = Jackfruit starch 70%: Chitosan 30%
P4 = Jackfruit starch 55%: Chitosan 45%

Procedures

Producing jackfruit skin flour uses a procedure that has been developed by Rizal et al. (2016) with modifications. The jackfruit skin is separated from the epidermis or rough skin, then sliced with a thickness of 0.5 mm. The jackfruit skin slices were washed using tap water, then soaked in a salt water solution with a ratio of 3:5 for 15 minutes. The jackfruit skin is washed using tap water and drained for 5 minutes. After that, the jackfruit skin is dried under in the sun for 7 days. Dry jackfruit skin in a blender until smooth, so that jackfruit skin flour is obtained.

Jackfruit Skin Extraction
Delignification process

Jackfruit skin starch as much as 100 g was weighed then added 10% NaOH 1000 mL with a ratio of 1:10 and refluxed at 80 °C or 1 hour 36 minutes. After that reflux is filtered to get solids, then washed and dried in the oven.

Bleaching Process

The result of the delignification process is then transferred to the bleaching process with a ratio of 1:10 using 30% hydrogen peroxide (v/v) at 90 °C for 1 hour. Furthermore, filtering is carried out, taking part of the residue. The residue obtained was washed with water to pH 7, then it is dried in the sun.

Producing bioplastics from jackfruit skin and chitosan with the addition of VCO as a plasticizer

Producing bioplastics from based on a research conducted by Shafqat A et al., (2021). The producing of bioplastics from jackfruit skin starch - chitosan is carried out in the following way: a solution of jackfruit skin starch and chitosan was mixed based on the determined ratios. Then VCO is added (10% of the addition of the total weight of starch solids and chitosan) and stirred for 1 hour until homogeneous. The resulting solution was then formed on a glass mold with a size of 20 cm x 15 cm and dried in an oven at 60 ^{o}C for 6 hours. The heated bioplastic was cooled at room temperature for 6 hours and then released from the glass plate and stabilized in a desiccator before being analyzed.

Observation Parameter

Bioplastics are characterized by the parameters of thickness, tensile strength, water absorption and biodegradability. Characterization of mechanical properties using the Brazilian Test (Chicago U-160-A). Test water absorption was conimmersing the bioplastic sample in 10 mL of water for 24 hours.

Bioplastic Thickness Test

The dried bioplastic was measured using a screw micrometer with an accuracy of 0.01 mm. According to Yao X et al. (2013) measurements were made at 5 different points on the bioplastic. Then the average thickness of the bioplastic was taken.

Tensile Strength

Tensile strength is based on the results of tensile strength tests by Tensile Strength ultimate. The research sample was in the form of bioplastic cut with a size 5 cm x 2 cm. The tensile strength value data obtained from the test results

$$.(MPa) = \frac{\text{Tengsile Strength}}{\text{Elongation}}$$

Water Absorption

Bioplastics were cut to a size of 3 cm × 3 cm. The cut bioplastic was then weighed using an analytical balance. After that, the bioplastic was put into a 100 mL beaker filled with 50 mL of distilled water, then allowed to stand for 2 minutes then removed and carefully dried using a tissue (Solekah et al., 2021). Bioplastics are weighed and their resistance to water is measured using the following equation calculated by the formula:

$$\text{Water Absorption (\%)} = \frac{(W-W_0)}{W} \times 100\%$$

where:
W_0 = Initial weight of sample
W = Sample final weight

Biodegradability

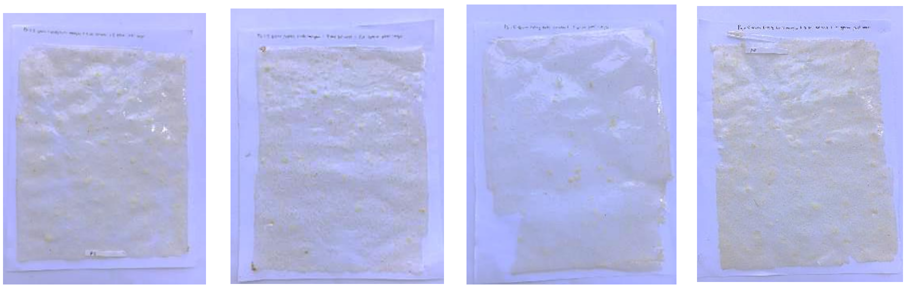
According to Nuryati, N (2019) the biodegradability test with the soil planted test method, the goal is to see the rate of sample degradation and how long the sample will be decomposed by microorganism in the soil. The research sample was in the form of bioplastic cut with a size of 4 cm x 1 cm. The sample is weighed then placed in a pot and observed every 3 days and weighed until a constant weight is obtained.

Data analysis

The ANOVA (Analysis of Variance) test was used to test the effect of each factor, followed by the Least Significant Difference BNT further test. The level of confidence used is 95% (α 0.05).

RESULT AND DISCUSSION

As a plasticizer, jackfruit skin starch is used in the production of bioplastic, along with chitosan and VCO (Figure 1). In addition, bioplastic characterization was performed, which included thickness tests, tensile strength tests, water absorption tests, and biodegradability tests.



P1 P2 P3 P4

P1 = Jackfruit Starch 100%: Chitosan 0% P2 = Jackfruit starch 85%: Chitosan 15%
P3 = Jackfruit starch 70%: Chitosan 30% P4 = Jackfruit Starch 55%: Chitosan 45%

Figure 1. Bioplastic of jackfruit cultivars with the addition of chitosan and VCO as a plasticizer

Characteristics of Bioplastic Thickness

The comparison of the optimum concentration of jackfruit skin starch: chitosan obtained in the thickness test of bioplastics, namely the ratio of jackfruit skin starch: chitosan of 55%:45%. In this ratio, the bioplastic has an elastic texture, formed.

Table 1. Thickness of bioplastics

Sample Skin Starch Jackfruit : Chitosan (%)	Average Value
100:0	3.27^{c}
85:15	2.16^{b}
70:30	2.43^{a}
55:45	2.52^{b}

Explanation: a.b.c "different letters in the notation column show different effects, namely at the level of $p<0.05$

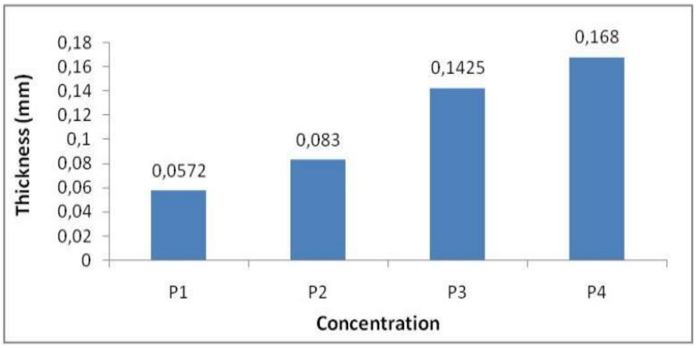


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Figure 2. Bioplastic thickness of jackfruit skin cellulose with the addition of VCO as a plasticizer

The results of the variance of bioplastic thickness showed that the ratio of jackfruit skin and chitosan in the producing bioplastics had a very significant effect with ($p < 0.05$). This is influenced by the addition of chitosan and VCO as a plasticizer. According to Anggraini et al. (2017) the increase in the concentration of chitosan affects the thickness of the bioplastic. The addition of glycerol from virgin coconut oil also has a significant effect on the thickness of bioplastics using the BNT test which shows the highest thickness value of 0.1682 mm at the addition of 10% VCO. The thickness of the bioplastic will increase with the addition of VCO due to the interaction of the glycerol molecule with the bioplastic polymer film (Hamzah HF et al., 2021). This is because glycerol or plasticizer has a low molecular weight so that it has the ability in the polymer matrix of protein and polysaccharides thereby increasing the formation and flexibility of the film (Sri Hastuti N, 2015).

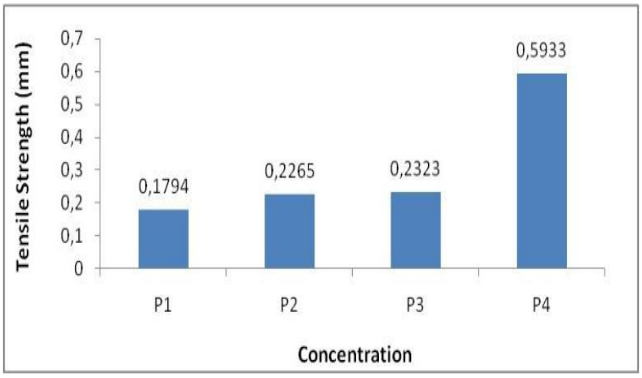
Tensile Strength

Tensile strength is carried out to determine the tensile strength of bioplastics in holding loads to protect products wrapped in bioplastics produced using different types and concentrations have different tensile strength values as shown in Table 2 and Figure 3.

Table 2. Tensile Strength

Sample Skin Starch Jackfruit : Chitosan (%)	Avarege Value
100:0	2.18^{c}
85:15	1.76.^{b}
70:30	3.23^{a}
55:45	1.82^{b}

Explanation: a.b.c "different letters in the notation column show different effects, namely at the level of p<0.05



P1= Jackfruit Starch 100% : Chitosan 0% P2 = Jackfruit starch 85%: Chitosan 15%
P3 = Jackfruit starch 70% : Chitosan 30% P4 = Jackfruit Starch 55% : Chitosan 45%

Figure 3. Bioplastic Tensile Strength

Figure 3 shows the tensile strength value for each ratio of jackfruit starch and chitosan in the range of 0.179-0.5933 MPa. The type and concentration of bioplastic constituents can affect the tensile strength value (Aditya Nandika A.J, et al., 2021). The results of the variance of bioplastic tensile strength showed that the ratio of jackfruit and chitosan in the producing of bioplastics had a very significant effect with (p <0.05). Bioplastics with the highest tensile strength values have a high affinity where the intermolecular bioplastics have a tendency to bond so as to produce strong bonds (Yefrita B et al., 2021). The highest tensile strength value of bioplastics in this study was in the research treatment of ratio 55%:45%, which was 0.5933 MPa, while the lowest tensile strength value is bioplastic in the research treatment of ratio 100%:0% which is 0.1794 MPa. The results of the tensile strength values start from the ratio of 85%:15% with 0.2265 MPa, to 0.2323 MPa in the ratio of 70%: 30% and peaked at 0.5933 MPa in the ratio of 55%:45%. The value of tensile strength increases with the addition of VCO because very closes the pores of the plastic film, thereby increasing the tensile strength of bioplastics. The higher the tensile strength value produced. The greater number of plasticizers used will increase the interaction between molecules in bioplastics so that the bonds between molecules in bioplastics are getting stronger (Sanyang L. M et al., 2015).

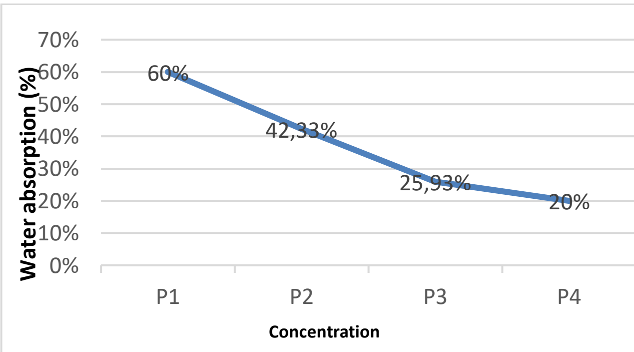
Water Absorption

Water absorption in bioplastics is used to determine the occurrence of bonds in the polymer and the level or regularity of the bonds in the polymer which is determined by the percentage addition of polymer (Abe M M et al., 2021).

Table 3. Water Absorption

Sample Skin Starch Jackfruit : Chitosan (%)	Avarege Value
100:0	3.19[c]
85:15	2.34.[b]
70:30	2..25[a]
55:45	1.47[b]

Explanation: a.b.c "different letters in the notation column show different effects, namely at the level of p<0.05



P1= Jackfruit Starch 100% : Chitosan 0% P2 =Jackfruit starch 85%: Chitosan 15%
P3 = Jackfruit starch 70% : Chitosan 30% P4 = Jackfruit Starch 55% : Chitosan 45%

Figure 4. Water Absorption Bioplastic

Figure 4 shows that the lowest level of water absorption is the ratio of jackfruit skin starch and chitosan concentration of 55:45% with water absorption of only 20% because the lower of water absorption in a bioplastic, the higher of water resistance, while the water absorption a very large amount will result in decreased water resistance and swelling in the sample. The results of the variance of bioplastic thickness showed that the ratio of jackfruit skin and chitosan in the producing of bioplastics had a very significant effect with (p <0.03. The highest water absorption is 60% which occurred in the ratio of jackfruit skin starch and chitosan concentration of 100%:0%. This ratio did not contain chitosan which will enable the plastic to decompose more easily, due to water absorption in the bioplastic (Foliono A et al., 2020).

The addition of chitosan causes the gel in the bioplastic to be tighter so that it does not absorb much water. This is due to the large structure of chitosan grains so that the water absorption capacity decreases (Czarnecka B et al., 2022).

Biodegradability

The biodegradability of bioplastics is an estimate of how long it takes for bioplastics to decompose properly in the environment.

Figure 5. Bioplastic Degradability

Bioplastic that has undergone a degradation process are visible changes in color and shrinkage (Mcrozkowska M et al., 2021). The color change occurs because the soil used is made moist. So that, the pores of the bioplastic absorb water and turn the bioplastic into white, initially clear. Some parts of the sample were destroyed because they were decomposed by bacteria in the soil and there was a decrease in weight for 3 days. This is because starch has a hydroxyl group (O-H) which initiates after absorbing water in the soil (Nanang E W et al., 2017). Starch polymers having hydroxyl groups OH will decompose into small pieces until they disappear in the soil.

Virgin Coconut Oil as a plasticizer also plays a role in degradation. Hydrophilic VCO donates OH groups to help absorb water in the soil. In this study at a ratio of 100:0% the initial weight 0.03 g and the third day became 0.01 g; ratio of 85%:15% the initial weight 0.08 g and on the third day became 0.04 g and ratio of 70%:30%, the initial weight 0.11 g and on the third day became 0.09 g.

Table 4. Percentage of Degradation Bioplastic

Sample Skin Starch Jackfruit : Chitosan (%)	Initial Weigth (g)	Final Weigth (g)	Percentage of Degradation (%)
100:0	0.08	0.04	50
85:15	0.11	0.09	18.18
70:30	0.09	0.03	66.66
55:45	0.06	0.03	50

Table 1 shows that the bioplastic treatment of ratio 100%:0% and ratio 70%:30% experienced the highest degradation at 66.66%. then the lowest degradation in sample the research treatment of ratio 85%:15% is 18.18%. Loss of weight indicates an indication of degradation in the soil. The bioplastic degradation test was carried out for 3 days and before planting the samples were weighed on the first day and then on the third day the final weight was also weighed. There was a decrease in sample weight on the third day.

Table 5. Bioplastic Breakdown Time

Sample Skin Starch Jackfruit : Chitosan (%)	Regression equation	Breakdown Time
100:0	y = -0.002x + 0.05	2,5
85:15	y = -0.04x + 0.12	3
70:30	y = -0.02x +0.13	6,5
55:45	y =-0.06x+0.15	2,5

Physical degradation occurs with a decrease in weight, color changes and brittleness in bioplastics. The length of time for bioplastics to fully degrade can be known by calculating the regression value for each sample. The value of x at y=0 indicates the time the bioplastic completely decomposes in the soil.

CONCLUSION

The addition of chitosan and virgin coconut oil (VCO) to the production of jackfruit skin starch bioplastics had a significant effect on thickness, tensile strength, water absorption, and biodegradability. The added amount of chitosan and VCO cause an increase in thickness, tensile strength, water adsorption and biodegradability of bioplastic. The ratio treatment of jackfruit skin starch and with a concentration of 55% and chitosan with a concentration of 45% is the best among study treatment.

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