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Optimization of Oil Palm Empty Fruit Bunches Cellulose-based Bioplastic Formulation with Response Surface Methodology (RSM)

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A B S T R A C T

Bioplastics are one of the alternatives to replace synthetic plastics. Bioplastics are plastics made from natural materials readily decomposed by microorganisms so that they are environmentally friendly. Oil Palm Empty Fruit Bunches (OPEFB) have cellulose content that can be utilized as raw material in the manufacture of bioplastics. This research aims to determine the best formulation of OPEFB cellulose-based bioplastics as environmentally friendly packaging. The method used in this research is the Response Surface Methodology (RSM) optimization method in the Mixture (Optimal Custom Design) section found in the Design Expert version 13 application. The factors analyzed in making bioplastics are glycerol, starch, CMC, and chitosan. Bioplastic observations include tensile strength, elongation, elasticity, thickness, water resistance, and biodegradation. The result of OPEFB cellulose obtained in this study is 71.88%, where the cellulose produces strong bioplastic properties. The optimum solution of bioplastic formulation produced is the addition of glycerol of 0.89 g, starch of 2.99 g, CMC of 2.11 g, and chitosan of 3 g Laboratory verification responses produced are tensile strength of 12 MPa, elongation of 31.34%, elasticity of 890.67 MPa, thickness of 0.25 mm, water resistance of 88.95%, and biodegradation of 4.34%/day.

Contribution to Sustainable Development Goals (SDGs):

SDG 9: Industry, Innovation and Infrastructure
SDG 12: Responsible Consumption and Production
SDG 13: Climate Action
SDG 14: Life Below Water
SDG 15: Life on Land
SDG 17: Partnerships for the Goals

1. INTRODUCTION

1.1. Research Background

Oil palm plantations in Indonesia continue to increase every year. Based on data from the Indonesian Ministry of Agriculture (MOA), the area of oil palm plantations in Indonesia will reach 16.83 million hectares (Ha) by 2023. This indirectly affects the waste generated from the palm oil industry, which is substantial waste in Oil Palm Empty Fruit Bunches (OPEFB). If the waste is not managed correctly, it will pollute the environment. OPEFB has a high cellulose content, so it has the potential to be utilized as an advanced material in the manufacture of bioplastics. Using cellulose from OPEFB as a raw material for bioplastics produces bioplastics with strong mechanical properties, meaning that the cellulose in the bioplastic matrix is in the form of fibers that function as reinforcement [1].

Several materials are needed to manufacture bioplastics, including a plasticizer for plasticizers and fillers. Glycerol is relatively effective in reducing internal hydrogen bonds, thereby increasing the intermolecular distance [2]. Adding glycerol plasticizer can increase the elongation of bioplastics made from OPEFB, but it weakens the tensile strength and does not improve water resistance and water vapor transmission rate [3]. These

weaknesses will be overcome by optimizing the formulation of the materials used.

Some other ingredients that can affect the biodegradable properties of bioplastics are starch, chitosan, and carboxy methyl cellulose (CMC). Adding starch in small amounts will produce thin films, and adding too much starch causes the film to become thick and difficult to print [4]. CMC can increase viscosity and tensile strength but decrease percent elongation. In addition, CMC can also increase ionic and chemical cross-linking to improve the mechanical properties of bioplastics [5]. Some studies suggest blending polysaccharides with CMC provides good mechanical and barrier properties due to the chemical similarity of the polysaccharides, thus allowing better compatibility [6]. Using chitosan is aimed at increasing the durability of bioplastics and has anti-microbacterial properties [6].

Given the many factors that determine the quality of bioplastics produced, it is necessary to conduct formulation optimization research on the manufacture of bioplastics by considering the variable percentage of cellulose, glycerol, CMC, starch, and chitosan. The rate of the addition of bioplastic forming components will greatly affect the quality and characteristics of the bioplastics produced. The percentage requirement of each composition can be calculated using the optimization formulation method.

1.2. Literature Review

1.2.1. Oil Palm Empty Fruit Bunches (OPEFB) Cellulose

OPEFB contains natural fibre material that is commonly used because it is cheap and non-toxic. It is a raw material in various applications such as power generation, paper industry, and composite formulations. Cellulose, lignin, and hemicellulose are the main components of oil palm solid waste, called lignocellulosic waste [7]. The main component of the dry weight of PKS is cellulose, up to 45% [8]. Cellulose is suitable as a plastic reinforcement material due to its abundant availability on earth. Its advantages include low cost, biocompatibility, non-toxic, edible, and environmentally friendly [9]. Cellulose is found throughout plant cell walls, including stems and wood. Some cellulose is soluble in alkaline solutions, but insoluble in organic solvents and water. Cellulose will be soft if it contains a lot of water and will be hard, brittle and hygroscopic in dry conditions [10].

1.2.2. Bioplastic Formulation

Bioplastics comprise several combinations of raw materials, including glycerol, starch, CMC, and chitosan. Glycerol as a plasticizer improves the plasticisation properties of plastics, including improving brittle and soft mechanical properties. The concentration of glycerol as a plasticiser will affect the value of elongation and tensile strength in the manufacture of bioplastics [11]. Starch consists of long chains of two types of glucose molecules: amylose, which has long straight glucose molecular chains, and amylopectin, which has short branched glucose molecular chains. In the production of biodegradable plastics, starch is one of the main ingredients of bioplastic production because it is elastic and similar to plastics made from petroleum polymers. The principle of gelatinization is used to form biodegradable plastics using starch as the base material.

The addition of CMC (Carboxy Methyl Cellulose) is also widely applied in the manufacture of bioplastics. CMC can also be used as a composite with starch because chemically CMC is relatively similar to starch [12]. In bioplastic production, adding CMC increases tensile strength as a filler material. Adding CMC to bioplastic production increases the tensile strength value, decreases the elongation rate, decreases water resistance, and accelerates the degradation of bioplastics produced [13].

The addition of chitosan can also increase the strength of bioplastics. Chitosan is a poly-amino saccharide formed through the deacetylation process of chitin. Bioplastic products show better mechanical properties and water resistance when using more chitosan [14]. Using chitosan as an additive also increases the transparency of the resulting plastic film [15]. Previous research on bioplastics still shows low mechanical properties (elastic modulus and tensile strength) and less resistance to water (hydrophilicity). This hydrophilicity can be reduced by adding chitosan [6].

1.2.3 Bioplastic Packaging

Bioplastics are biopolymer compounds that are naturally degraded by fungi, bacteria, and algae or undergo hydrolysis. Bioplastics have been developed more than 10 years ago, and the development is considered slow. This is due to their high technology and cost and their different properties from conventional plastics. However, along with reducing petroleum reserves, bioplastics have become a competitive product compared to other plastics [16]. Bioplastics have properties that are flexible, odourless, easy to print, can inhibit the entry and exit of gas and water vapour, transparent and non-toxic when burned or do not become garbage because they are environmentally friendly.

Based on the description above, many researchers are currently trying to protect the environment and look for other alternatives as raw materials for bioplastics. One of these efforts is to produce plastics from natural raw materials, which are compounds that are environmentally friendly and easily or quickly decomposed by microorganisms. Natural raw materials available in nature can be the basic ingredients of bioplastics because they have non-toxic and renewable properties. These renewable materials have high biodegradability so that they have the potential to be used as raw materials for making bioplastics [17].

1.3. Research Objective

This study aims to examine the effect of the addition of starch, glycerol, chitosan, and CMC in different treatments on the characteristics of bioplastics produced, as well as to obtain formulation optimization conditions in the manufacture of bioplastics made from OPEFB cellulose.

2. MATERIALS AND METHODS

2.1 Research Design

The research design in this research method uses a formula optimization technique using Response Surface Methodology (RSM) with a Mixture Optimal Custom Design (OCD) design contained in Design Expert 13 software. The variables in this

design consist of 4 factors, namely glycerol, starch, CMC, and chitosan. The response values were tensile strength, elongation, elasticity, thickness, water resistance, and biodegradation.

Based on the research in the manufacture of cellulose-based bioplastics from seaweed waste with a range of glycerol additions (0% - 1%), tapioca starch (2% - 10%), CMC (1% - 5%), and chitosan (1% - 5%) produced the best bioplastic characteristics with the addition of 0.165% glycerol, 3.78% starch, 2.5% CMC, and 1.62% chitosan [6]. From this value, the lower and upper points of the percentage are taken with the reference value being the best. The results of the design with OCD can be seen in Table 1.

Table 1. Treatment Combinations by the Optimal Experimental Design (Custom) Design for the Bioplastic Manufacturing Process from OPEFB

Run	A: Glycerol (g)	B: Starch (g)	C: CMC (g)	D: Chitosan (g)
1	0.5	2	3.5	3
2	1	3.5	1.5	3
3	0.5	4	1.5	3
4	0.75	4.84	1.5	1.90
5	0.75	4.84	1.5	1.90
6	0.5	4.40	2.92	1.17
7	1	3.41	2.25	2.33
8	1	3.02	3.5	1.47
9	0.5	3.98	2.62	1.88
10	0.5	2	3.5	3
11	0.64	3.85	3.5	1
12	0.79	2.70	2.59	2.9
13	1	2.3	3.5	2.2
14	0.5	6	1.5	1
15	0.5	4.25	1.95	2.28
16	0.75	4.84	1.5	1.90
17	0.5	3.98	2.62	1.88
18	0.79	2.70	2.59	2.9
19	1	5.23	1.63	1.12
20	0.79	2.70	2.59	2.9

2.2. Delignification of OPEFB

The delignification process of the OPEFB fibre was carried out to remove the lignin in the OPEFB fibre which hinders cellulose retrieval. The delignification process was carried out by mixing 20 g of OPEFB fibre with 0.5 M NaOH solution in a ratio of 1:10. The OPEFB fibre and NaOH were put into a 250 ml Erlenmeyer and covered using cotton and aluminum foil. After mixing, the sample was placed into the microwave with a heating time of 25 minutes at medium-high temperature. The results of delignification then carried out the bleaching process using 2% H2O2 with the ratio of H_2O_2 to distilled water is 1:15. Furthermore, it was heated using a hot plate for 2 hours at 60°C. The bleaching results were then washed using distilled water until the pH was neutral and dried using an oven at 100°C for 6 hours [18].

2.3 Manufacture of Bioplastics

Cellulose from long fibre fraction OPEFB was crushed using a hammer mill with a rotation speed of up to 18000 rpm for ± 5 minutes. Then sieving was carried out using a 100 mesh sieve. Tapioca starch (according to treatment) and CMC (according to treatment) were dissolved with 30 ml distilled water using a hot plate at 80°C for 15 minutes until gelatinization. Chitosan (according to treatment) was dissolved with 20 ml distilled water and heated using a hot plate for 5 minutes until the solution was homogeneous. The starch and chitosan solution was put into a 250 mL beaker glass, then 1 g of cellulose powder, glycerol (according to treatment), and 50 ml of distilled water. The mixture was heated on a hot plate at 70°C and stirred using a magnetic stirrer at 320 rpm for 30 minutes. The solution was then printed on a 17 x 9 cm glass plate and dried using a food dehydrator at 50°C for 12 hours. The resulting bioplastics were observed for their physical, mechanical, and biodegradation characteristics.

2.4 Analysis Procedure

2.4.1 Tensile Strength, Elongation, and Elasticity Tests (ASTM D-882)

Tensile strength testing uses a tensile tester where the sample is formed according to the ASTM D-882 standard size.

a. The tensile strength can be calculated using the following equation:

$$\sigma \quad \sigma = F/A$$

Description:
σ = Tensile strength (N/mm^2)
F = Pulling force (N)
A = Area of bioplastic film (mm^2).

b. Elongation is calculated using the following equation:

$$(\%) = (Pf-Pi)/Pi \times 100\%$$

Description:
e = elongation (%)
Pf = final length of the sample (mm)
Pi = initial length of sample (mm)

c. Modulus young is calculated using the following equation:

$$E = (\sigma) / e$$

Description:
E = Modulus young
σ = Tensile strength (N/mm^2)
e = Elongation (%)

2.4.2 Thickness (ASTM, 2005)

Thickness testing was carried out using a screw micrometer (accuracy 0.01 mm) by taking measurements at five different points, namely the upper right corner, lower right corner, upper left corner, lower left corner, and centre. The average calculation was calculated using the following equation:

Average thickness= ((point 1+point 2+point 3+point 4+point 5))/5

2.4.3 Water Resistance Test

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Weighing was carried out to determine the initial weight of the plastic, and then 2x2 cm plastic film was put into a glass that had been given 50 ml of aquadest. Immersion was carried out for 20 minutes at room temperature; after 20 minutes, the biodegradable plastic was taken, and then the plastic sample pieces were dried with tissue. Calculation of water resistance can be calculated using the following equation:

% Solubility = (Wet sample weight (g)- dry sample weight (g))/(dry sample weight (g)) X 100%

The value of water resistance can be calculated using the following equation:

Water resistance = 100% - % solubility

2.4.4 Biodegradability Test

The biodegradability test was conducted by weighing a 3 x 2 cm film sample, then placing the sample in a container filled with soil and left for several days. Observations were made over nine days. The weight loss of the biodegradable plastic was calculated using the following equation:

Weight loss = (W0-W1)/W0 x 100%

Descriptions:
W0 = Initial Weight Before Burial (g)
W1 = Final Weight After Burial (g)

The biodegradability of a material can be known by testing the percentage of weight gained over time. Biodegradability is determined using the following equation:

Biodegradability= (Weight loss)/time

3. RESULT AND DISCUSSION

3.1. Chemical Characteristics of Raw Materials

Cellulose from OPEFB is obtained by delignification, which is a process that breaks down lignocellulosic materials into cellulose, hemicellulose, and lignin. Based on previous research OPEFB, before delignification, contains 34.06% cellulose, 14.42% hemicellulose, and 40.83% lignin [19]. High lignin content is one of the inhibiting factors in cellulose utilization. This OPEFB delignification research results produced 71.88% cellulose, 0.90% hemicellulose, and 19.92% lignin. The cellulose content obtained is higher than that obtained by the other research, which produced a cellulose content of 42.7% using a digester [20]. The cellulose content increased because the microwave worked by passing microwaves to the material/OPEFB. Microwave absorption causes a very rapid increase in temperature in reactants, solvents, and products. The microwave interaction with the material causes the hemicellulose content that binds cellulose to be released, and the lignin content in the cell wall that blocks cellulose begins to decrease [18].

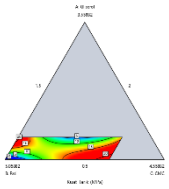
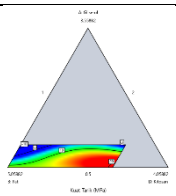
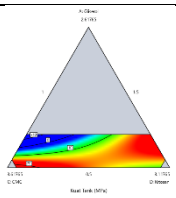
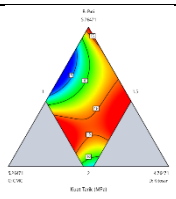
The cellulose obtained from the delignification process is then subjected to a bleaching process. The bleaching process aims to remove pigments and residual lignin and simultaneously bleach the cellulose from the delignification process. According to other research, a bleach that reacts with lignin contained in OPEFB can break the bonds on the O atom. The O atom binds to the C element contained in the benzene ring in the lignin compound [21]. When the bonds of the O atoms release other bonds and bind to the benzene ring, the lignin is reduced, and its size becomes smaller and even disappears. Cellulose, after bleaching, will produce a lighter color. This light color indicates that the pigments and lignin have dissolved so that high-purity cellulose is obtained [22].

3.2 Surface Response Analysis of Bioplastics

3.2.1 Analysis of Tensile Strength Values

Tensile strength is the maximum pull achieved until the plastic can remain before breaking. Based on the test results that have been carried out, the range of values obtained ranges from 3.12 MPa to 16.4 MPa, as can be seen in Table 2. |

Table 2. Interaction and Tensile Strength Response Contour Graphs

Inter-action	Countour Graph	Descriptions
ABC		The highest response is shown in red in the middle of variables B and C. The lowest response value is blue on variables A and B. The highest predicted tensile strength response from ABC interaction is 20 MPa.
ABD		The design expert graph with the highest response value is red, which is in the middle of variables B and D. The lowest response value is blue, which is between variables A and B. The highest predicted tensile strength response from the ABD interaction is 20 MPa.
ACD		The highest response value is shown by the red color in variable C and between variables A and D. The lowest tensile strength response is in variable A. The highest predicted tensile strength response of the ACD interaction is 20 MPa.
BCD		The red color shows the highest response value between variables B, D, and C. The lowest variable is in variables B and C. The highest predicted tensile strength response of the BCD interaction is 15 MPa.

The model suggested by the program for the response to the results of this study is the unique cubic model. This model was chosen because it has the lowest PRESS (prediction error sum of square) value compared to other models. The prediction from the sequential model sum of the squares shows that the p-value <0.05 is 0.0085 for tensile strength. This means that the special cubic experimental model is statistically significant.

Based on the interaction of ABC and ABD, the tensile strength value increases by adding CMC and chitosan. The higher the addition of CMC and chitosan, the tensile strength will increase. The presence of cellulose components and the addition of CMC tend to increase the tensile strength of bioplastics. In addition, the increase in tensile strength is also due to a suitable interface adhesion force where a strong hydrogen bond is formed between the hydroxyl group (O-H) of starch and the hydroxyl group (O-H) of cellulose. Cellulose fiber will reduce the pore space in the plastic, making it stronger and more resistant to pulling [23]. Adding chitosan as a filler in the manufacture of bioplastics can also increase hydrogen bonds so that bioplastics become more muscular and not easily broken [24].

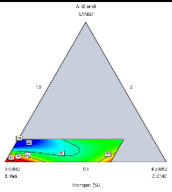
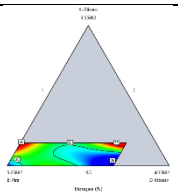
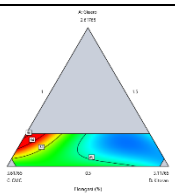
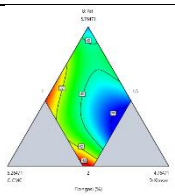
The addition of glycerol and starch in the manufacture of bioplastics can decrease the tensile strength, as seen from the interaction of ABD. This is to the research stated that increasing glycerol can decrease the value of tensile strength and modulus young because glycerol can reduce the internal hydrogen bonds of molecules, thereby weakening the attraction of adjacent polymer chain intermolecular [25]. Based on the Indonesian National Standard (SNI) 7188.7 of 2016, bioplastics must have a tensile strength value of 24.7 to 302 MPa. The research results obtained a tensile strength value of 3.12 to 16.44 MPa, meaning it has not met the Indonesian National Standard. However, according to the Japanese International Standard (JIS) in 1975, the minimum tensile strength value is 3.92 MPa, and the results of this study show that the value meets the minimum standard.standard.

3.2.2 Analysis of Elongation Value

An elongation or elongation test at break is the ability of a material to stretch when pulled. Elongation aims to determine the elasticity of a bioplastic film. The bioplastic elongation response values from the test results ranged from 28.06% to 33.76%. The model suggested by the program for the response to the results of this study is the special cubic model. The prediction of the sequential model of the sum of the square model shows that the p-value < 0.05 is 0.0320 for elongation. This number indicates that the model is statistically significant. The interaction of the elongation response can be seen in Table 3.

Adding glycerol, starch, CMC, and chitosan affected the elongation response differently. Adding glycerol as a plasticizer can reduce the force between molecules, which causes the polymer chain bonds to become freer so that bioplastics will be more flexible [26]. The increasing addition of chitosan causes the elongation value to decrease. This happens because adding chitosan can cause the bioplastic film to become stronger and stiffer so that the resulting plastic becomes rigid. The amount of chitosan added can decrease the bond distance between molecules [27].

Table 3. Interaction and Elongation Response Contour Graph

Inter-action	Countour Graph	Descriptions
ABC		The red color in variable B indicates the highest response. The blue color between variables A and B indicates the lowest response value. The highest elongation response prediction from the ABC interaction is 36%.
ABD		The highest response value is in variable A and between variables A and D. The lowest response value is blue in variables B and D. The highest predicted elongation response from ABD interaction is 35%.
ACD		The highest response value is red between variables A and C, while the lowest elongation response is between variables A and D. The highest elongation response prediction of ACD interaction is 36%.
BCD		The red color shows the highest response value between variables B and C and C and D. In contrast, the lowest variable is located in variables B and D. The prediction of the highest elongation response from BCD interaction is 34%.

The interaction between starch, CMC, and chitosan also resulted in a decrease in elongation value. The addition of tapioca starch as a filler can reduce the elongation value. Starch in large quantities can produce bioplastic films that are increasingly brittle or brittle so that the elongation percentage becomes low [28]. The increase in elongation value is inversely proportional to the tensile strength. Increased elongation occurs because the distance between molecules gets farther, so the bonding chain becomes freer and flexibility increases. Based on the Indonesian National Standard (SNI) 7188.7 of 2016, bioplastics have a ductile value of 20-220%, meaning that the bioplastics produced from this study have met the SNI.

3.2.3 Analysis of Modulus Young Value

The elasticity test or modulus young parameter shows the bioplastic film's stiffness level [29]. The results of the elasticity value test showed a value range between 831.85 MPa to 2438.8 MPa. The model prediction obtained is a special cubic, which shows that the p-value < 0.05 is 0.0061, so the model is statistically significant. The interaction of elasticity response can be seen in Table 4.

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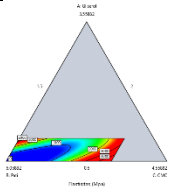
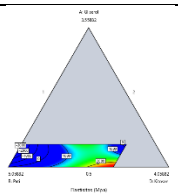
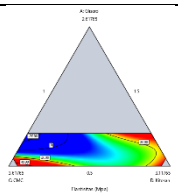
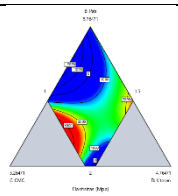
Table 4. Interaction and Modulus Young Response Contour Graph

Inter-action	Contour Graph	Description
ABC		The red color indicates the highest response in variable C and between variables A and B. The blue color in variable B indicates the lowest response value. The highest elasticity response prediction of the ABC interaction is 4000 MPa.
ABD		The highest response is indicated by the red color in the middle of variables B and D. The blue color in variables A and B indicates the lowest response value. The prediction of the highest elasticity response of the ABD interaction is 2000 MPa.
ACD		The highest response value is red between variables A and D and variable C, while the lowest elasticity response is in variable A. The highest elasticity response prediction of ACD interaction is 4000 MPa.
BCD		The red color shows the highest response value between variables C and D, while the lowest is in variable B. The prediction of the highest elasticity response of the BCD interaction is 2000 MPa.

Adding glycerol, starch, CMC, and chitosan affects the elasticity value of bioplastics. In the study results, the highest elasticity value obtained was 2393.4 MPa, while the lowest was 832.65 MPa. The high elasticity value is due to the relatively small addition of glycerol, resulting in a relatively small elongation value and a high elasticity/modulus young value [29]. In the ABD interaction seen on the contour graph, chitosan and glycerol produce high elasticity values due to the chemical interaction between chitosan and other molecules, such as glycerol, which can increase overall elasticity and mechanical strength. This interaction can improve the molecular mobility of polymer chains so that elasticity rises [30].

3.2.4 Analysis of Thickness Value

The bioplastic film's thickness is an important parameter affecting plastic use as packaging. Based on the results of the tests that have been carried out, the range of thickness values obtained is 0.24 mm to 0.27 mm. The model suggested by the program for the response of the results of this study is the quadratic model. The model prediction obtained from the sequential model sum of squares shows that the $p<0.05$ value is 0.0428, so it can be said that the quadratic experimental model is statistically significant. The interaction between factors (glycerol, starch, CMC, and chitosan) in the manufacture of bioplastics can be seen in Figure 1.

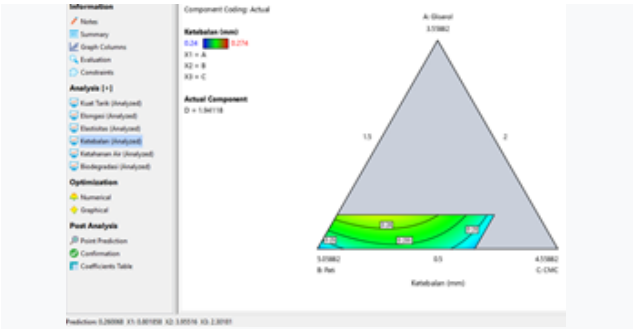


Fig 1. Thickness of Countour Graph

The blue color indicates the low thickness response, while the yellow color indicates the higher elasticity value. The composition of the raw materials used affects the bioplastic film's thickness. Based on the contour graph in Figure 1, it can be seen that the thickness values are almost the same for each treatment. This is because the total composition of the material is the same, which is 9 g, and the more raw materials are added, the more the thickness value also increases. The thickness of the bioplastic film is also affected by the content of the dissolved fraction, the area of the mold, and the volume of the solution in it. This is based on research conducted by other researchers, which states that the amount of solution used also affects the thickness level of bioplastics. The higher the thickness of bioplastics, the more rigid and hard the bioplastics will be, so the value of tensile strength and elongation will decrease because bioplastics break quickly [24]. Based on the JIS 2-1707 1946 standard, the maximum value for bioplastic thickness is 0.25 mm, and the result of the average thickness of bioplastic according to the contour chart shows that the test value has been qualified.

3.2.5 Analysis of Water Resistance Value

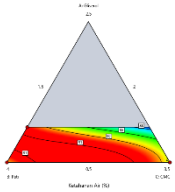
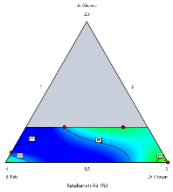
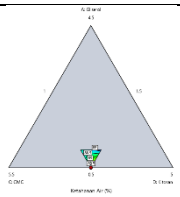
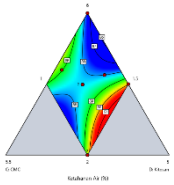
Water resistance or swelling test is the biodegradable development level in water absorption. Bioplastics are expected to have good water resistance properties because one of the properties of plastic is hydrophobic or waterproof so plastic can be wider for primary and secondary packaging. Based on the results of the tests that have been carried out, the range of water resistance values obtained is 87.54% to 90.52%. The suggested model for this response is a linear and unique cubic model and is statistically significant ($p<0.05$ i.e., <0.0001). The interaction and graph of water resistance contours can be seen in Table 5.

The interaction in the ABC and BCD graphs shows that the water resistance of bioplastics tends to decrease with the addition of CMC. This was achieved by the other research, which obtained a lower water resistance value along with an increase in the concentration of glycerol and CMC fillers [31]. The more CMC is added to the manufacture of bioplastics, the easier it will be to absorb water, which will cause the water resistance to be lower. CMC is hydrophilic, making water quickly enter the bioplastic matrix and causing bioplastics to be easily damaged or brittle. [32].

The use of chitosan in the manufacture of bioplastics can increase the water resistance of bioplastics. This is evident in this study, which can be seen in the interaction of ABD and BCD graphs, which show that water resistance increases with adding chitosan. This is because chitosan is hydrophobic (does not like

water). The resistance of bioplastic films to water is influenced by the chitosan fraction; that is, water resistance will be better if the chitosan fraction is more significant, namely with water uptake having the best value. Based on the Indonesian National Standard (SNI) 7188.7 of 2016, the maximum water security value is 99%. This study's water resistance value has almost met the national bioplastic standard.

Table 5. Interactions and Water Resistance Response Contour Graphs

Inter-action	Contour Graph	Description
ABC		The red color indicates the highest response in variables B to C. The blue color between variables A and C indicates the lowest response value. The highest prediction of water resistance response from the ABC interaction is 91%.
ABD		The ABD interaction showed the highest value, with the green color being in variables A and D. The lowest response value was indicated by the blue color between variables A and B. The highest prediction of water resistance response from the ABD interaction was 89%.
ACD		The highest response value is green, between variables A, C, and D. The highest prediction of water resistance response from ACD interaction is 89.6%.
BCD		The red color shows the highest response value in variable D. The lowest variable is located in variables B and C. The highest prediction of water resistance response from BCD interaction is 91%.

3.2.6 Biodegradable Value Analysis

Biodegradability is the biochemical decomposition process of organic molecules by microorganisms that convert C, N, S, and P (organic compound content) into inorganic products [32]. The biodegradation response values of bioplastics tested ranged from 3.97%/day to 4.89%/day. The model suggested by the program for the response of the results of this study is the linear model. The model prediction obtained from the sequential model sum of squares shows that the p-value < 0.05, which is 0.0043, means the model is statistically significant. The blue color indicates the low biodegradability response, while the higher elasticity value is indicated by the yellow color, which can be seen in Figure 2.

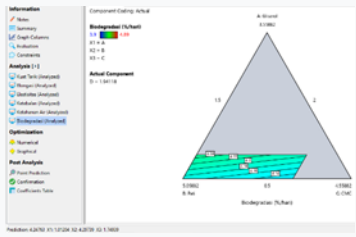


Fig 2. Biodegradation of Contour Graph

The crystallinity of cellulose is reduced due to the addition of glycerol, CMC, and chitosan, which can accelerate the biodegradation process of bioplastic films. Using starch as a matrix and glycerol as a plasticizer can affect the biodegradation of plastics. The OH groups in glycerol and starch will be hydrolyzed with water, decomposing the bioplastic into smaller sizes and eventually disappearing from the soil [14]. Chitosan as a filler has hydrophobic properties that are insoluble in water, so the degradation ability will be inhibited by adding chitosan [33]. It can be seen in the contour graph that the highest biodegradability value occurs in the addition of glycerol and starch. This is because the glycerol-starch mixture in bioplastics forms weaker hydrogen bonds, so bioplastics decompose faster. The use of cellulose can also affect the rate of plastic biodegradation. The higher the addition of cellulose concentration, the higher the biodegradation rate also increases [30]. Based on SNI No. 7188.7 of 2016, the duration of biodegradation ability for bioplastics is within 60 days to be able to decompose in its entirety. The duration of the degradation ability resulting from this study is 3-4%/day, which means that it will be degraded in the range of 25 to 33 days. The degradable ability has met the set bioplastic standards.

3.3 Surface Response Optimization

Optimization aims to find the best combination of models that produce the desired response and variables. Optimization was carried out to obtain the best combination of treatment between several factors (glycerol, starch, CMC, and chitosan) to the characteristics of the resulting bioplastics, namely tensile strength, elongation, elasticity, thickness, water resistance, and biodegradation. The method used is the multiresponse method, also known as desirability. The range of desirability values is between 0 and 1, with the best optimum value indicated by a desirability value close to one. In Table 5, the target of the component (goal), the weight of the importance of each variable, and the response to get a solution from the variable that produces the optimal response can be known. The description of the variables and responses to be optimized from this study can be seen in the following Table 6.

Variables such as glycerol, starch, CMC, and chitosan were optimized by setting a component target (goal) within the specified range with a positive importance weight of 3 (+++). This is due to the possibility that the optimal response is not achieved at the central point but rather at the recommended values. The response to tensile strength, elongation, water resistance, and biodegradation is set to maximize obtaining good mechanical characteristics of bioplastics. At the same time, the thickness is placed at the goal in range to find out the best thickness value from the results of the research that has been carried out. The best solution produced by Design-Expert can be seen in Table 7.

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Table 6. Description of Variables and Responses to be Optimized

Criteria	Goal	Lower limit	Upper limit	Importance weight
Glycerol	in range	0.5	1	3 (+++)
Starch	in range	2	6	3 (+++)
CMC	in range	1.5	3.5	3 (+++)
Chitosan	in range	1	3	3 (+++)
Tensile strength	maximize	3,12	16.44	3 (+++)
Elongation	maximize	28.06	33.76	3 (+++)
Modulus young	minimize	117	2438.8	3 (+++)
Thickness	in range	0.24	0.274	3 (+++)
Water resistance	maximize	87.54	90.52	3 (+++)
Biodegradations	maximize	3.97	4.89	3 (+++)

Table 7. The Best Solution for Analysis Results

Glycerol	Starch	CMC	Chitosan	Tensile trength	Elongation	Modulus young	Thickness	Wtaer resistance	Biodegradability
0.89	2.99	2.11	3.0	11.1	32.3	998.7	0.26	90.2	4.45
0.87	2.91	2.21	3.0	10.9	32.1	874.3	0.26	90.2	4.45
0.92	3.29	1.79	3.0	13.2	32.2	1508.1	2.26	90.5	4.51

Based on the optimization solution in Table 7, the most optimal solution is obtained in row one. Therefore, labor verification is carried out to ensure that the research results are highly accurate and reliable. The results of laboratory verification can be seen in Table 8.

Table 8. Laboratory Verification

Glycerol	Starch	CMC	Chitosan	Tensile strength	Elongation	Modulus young	Thickness	Water resistance	Biodegradability
0.89	2.99	2.11	3.0	12,0	31,34	890,67	0,252	88,95	4,34

After the labor verification, the results were not significantly different from the optimum solution. The optimal selective treatment is the first-line solution treatment indicated by the software, and several factors influence it. The treatment in the first solution was chosen because the results of laboratory verification were not much different from the optimal solution.

4. CONCLUSION

Using cellulose made from OPEFB as a raw material for making bioplastics results in more substantial bioplastic characteristics. Adding glycerol, starch, CMC, and chitosan affects the tensile strength, elongation, elasticity, water resistance, thickness, and biodegradation value. Adding glycerol can increase the elongation and biodegradation value and decrease the tensile strength, elasticity, and water resistance. Adding starch in large quantities can reduce tensile strength, elongation, elasticity, and water resistance and accelerate biodegradation. Adding CMC and chitosan can improve tensile strength and elasticity, lower elongation value and water resistance, and accelerate biodegradation. The results of the optimization of the process of making bioplastics from OPEFB cellulose with variations in the addition of glycerol, starch, CMC, and chitosan with the recommended optimum conditions are a combination of glycerol 0.89 g, starch 2.99 g, CMC 2.11 g, and chitosan 3 g. The response values produced from laboratory verification were tensile strength of 12 MPa, elongation of 31.34%, elasticity of 890.67 MPa, thickness of 0.25 mm, water resistance of 88.95%, and biodegradation of 4.34%/day.

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