Optimization of physical characteristics of bioplastics from agricultural waste using response surface methodology (RSM)

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Abstract. Rapid technological advances are in line with the increasing use of plastics. The problem faced is that plastic waste is not easily degraded by microorganisms in the soil. Bioplastics are made from renewable natural materials so that they have properties that can be broken down by microorganisms. In this study, bioplastics were made from agricultural waste containing starch, namely from jackfruit seed starch plus cornocob starch. Optimization using the response surface methodology (RSM) was carried out to see the optimum conditions of the variables and also the response to the physical properties of the resulting bioplastic. The research variables used were ZnO consisting of 3, 6, and 9 percent (%) and the glycerol variable consisted of 2, 5, and 8 mL. From the test results using RSM obtained the optimum conditions of ZnO 7.42% and glycerol 4.96 mL with a residual value of 81.3612%; water resistance 86.1333%; and heat resistance of 96.888°C with a desirability value of 0.725. The combination of the basic ingredients of agricultural waste in the form of jackfruit seeds and corn cobs is able to produce bioplastics with good physical properties.

1. Introduction

Rapid technological advances are in line with the increasing use of plastics. No wonder the increasing pile of waste in the form of plastic as a result of its increasing use. There is a lot of garbage that is deliberately dumped into the river so that it can clog waterways. The problem faced is that plastic waste is very difficult to decompose by microorganisms in the soil, thus making it accumulate in the soil in increasing numbers and ultimately causing environmental pollution. To overcome this, it is often found where the waste in the form of plastic is destroyed by burning, so that it can produce carbon emissions that are very dangerous for the surrounding environment. Based on data from the Indonesian Aromatic and Plastic Olefin Industry Association (INAPLAS), in 2015 plastic consumption in Indonesia reached 17 kg/capita/year. Based on these data, it can be concluded that if there was an increase in the population of Indonesia in the first half of 2017 of 261 million people, the national use of plastic could reach 4.44 million tons [1]. The type of plastic that is commonly used is derived from synthetic polymers from petroleum. The disadvantage of plastics with synthetic polymer materials is that they are limited in number and difficult to recycle [2].

Hazardous compounds produced from open burning include CO, CO₂, CH₄, SO₂, and N₂O gases produced from the combustion process are categorized as greenhouse gases that can cause global warming [3]. Other materials can have a negative impact on health in the short and long term. A solution is needed to be able to reduce the occurrence of environmental problems caused by the presence of...
plastic waste, one way that can be done is by replacing conventional plastic materials into environmentally friendly plastic materials (bioplastics) that can be decomposed in the soil. Bioplastic is a type of plastic made from natural materials that can be renewed so that it has properties that can be decomposed by microorganisms [4]. Indonesia has potential in the development of bioplastics, this is supported by agricultural and marine products that can be developed into biopolymers as materials for making bioplastics [5]. Bioplastics can be made from agricultural materials containing polymeric compounds such as starch, cellulose and lignin, and can also be derived from animals such as casein, chitin and chitosan and so on [6]. The advantages of using cellulose in the manufacture of bioplastic, among others, are easy to obtain, usually used as a reinforcing material in the manufacture of biodegradable plastics [7]. In this study, the manufacture of bioplastics from agricultural waste that contains starch, namely from jackfruit seed starch plus corn cob starch. This type of waste is mostly disposed of after the main benefits have been obtained, thus turning it into unused waste. However, by turning it into a raw material in the manufacture of bioplastics, it can directly overcome the troubling problem of plastic waste and at the same time make the waste a material that has useful value.

In this study, Optimization using RSM was carried out to obtain the optimum point on the variable and also the response in the form of physical properties of bioplastics.

2. Materials and methods
2.1. Materials
Some of the materials used include corn cob starch and jackfruit seeds, glycerol, ZnO, water, soil, acetic acid, trays, scales, label paper, thermometers, stoves, blenders, and molds with size 13 x 13 centimeter.

2.2. Methods
This study uses a factorial design where the first factor is ZnO which consists of 3, 6, and 9 %. The second factor is glycerol which consists of 2.5 and 8 mL. The response that will be seen in this study is in the form of physical characteristics of bioplastic consisting of % residual, % water resistance, and % bioplastic resistance to heat. [8] By using the RSM method with software DX 7.0.0, 13 treatment combinations will be obtained.

3. Results and discussion
Based on the results of research using the RSM method with software design expert 7.0.0, the physical properties of bioplastics are obtained in the form of % residual, % water resistance, and % heat resistance of bioplastic which can be seen in Table 1 below.
### Table 1. Response physical characteristics of bioplastics.

| Run | ZnO (Coded) | Glycerol (Coded) | ZnO (%) | Glycerol (%) | Residual (%) | Water Resistance (%) | Heat Resistance (°) |
|-----|-------------|------------------|---------|--------------|--------------|----------------------|---------------------|
| 1   | 0           | -1.4142          | 6       | 0.76         | 92.176       | 96.976               | 94.0                |
| 2   | 0           | 1.4142           | 6       | 9.24         | 76.049       | 70.333               | 83.0                |
| 3   | -1.4142     | 0                | 1.76    | 5            | 81.392       | 74.540               | 68.0                |
| 4   | 1.4142      | 0                | 1.024   | 5            | 85.13        | 82.950               | 98.0                |
| 5   | -1          | 1                | 3       | 8            | 76.479       | 71.077               | 70.0                |
| 6   | 1           | -1               | 9       | 2            | 90.5         | 80.89                | 97.5                |
| 7   | 1           | 1                | 9.24    | 8            | 75.979       | 77.273               | 95.0                |
| 8   | -1          | -1               | 3       | 2            | 88.98        | 87.571               | 81.0                |
| 9   | 0           | 0                | 6       | 5            | 80.85        | 86.538               | 93.5                |
| 10  | 0           | 0                | 6       | 5            | 80.818       | 85.300               | 94.0                |
| 11  | 0           | 0                | 6       | 5            | 81.718       | 85.320               | 92.0                |
| 12  | 0           | 0                | 6       | 5            | 79.645       | 86.615               | 93.0                |
| 13  | 0           | 0                | 6       | 5            | 80.2500      | 84.714               | 92.5                |

Source: Data from research results

### Table 2 ANOVA respon of physical characteristics of bioplastic

| Source                     | Residual (%) | Water Resistance (%) | Heat Resistance (°) |
|----------------------------|--------------|----------------------|--------------------|
| Prediction models          | Quadratic    | Quadratic            | Quadratic          |
| Model                      | <0.0001      | <0.0001              | <0.0001            |
| A-ZnO (%)                  | 0.0734       | 0.0004               | <0.0001            |
| B-Glycerol (ml)            | <0.0001      | <0.0001              | <0.0001            |
| AB                         | 0.3723       | 0.2712               | 0.0004             |
| A²                         | 0.0263       | 0.0002               | <0.0001            |
| B²                         | 0.0062       | 0.1249               | <0.0001            |
| Lack of fit                | 0.1532       | 0.0542               | 0.7889             |
| Std. Dev                   | 1.06         | 1.2                  | 0.67               |
| Mean                       | 82.31        | 83.01                | 88.58              |
| C.V. %                     | 1.29         | 1.45                 | 0.76               |
| PRESS                      | 42.7         | 62.32                | 8.65               |
| R-Squared                  | 0.9773       | 0.9859               | 0.9974             |
| Adj R-Squared              | 0.9612       | 0.9758               | 0.9955             |
| Pred R-Squared             | 0.877        | 0.9133               | 0.9928             |
| Adeq Precision             | 24.471       | 32.153               | 64.933             |

Source: Data processing results (5%)

### 3.1. Analysis response % residual

Based on the results of data processing, the equation for the % residual is in the form of a quadratic which can be written as follows:
Based on the data in Table 2, it can be seen that the variables that affect the % residual response are $X_2$ (glycerol), $X_1^2$ (ZnO squared), and $X_2^2$ (the square of glycerol). From equation (1), the eigenvalues of the matrix are both positive, namely $\lambda_1 = 1.89$ and $\lambda_2 = 0.78$, so that the resulting surface response is a minimum response [9]. The response surface model can be seen in the following figure:

$$Y = +96.59742 - 0.95913X_1 - 3.46464X_2 - 0.056111X_1X_2 + 12.520X_1^2 + 0.17251X_2^2$$  \hspace{1cm} (1)$$

Based on the data in Table 2, it can be seen that the variables that affect the % residual response are $X_2$ (glycerol), $X_1^2$ (ZnO squared), and $X_2^2$ (the square of glycerol). From equation (1), the eigenvalues of the matrix are both positive, namely $\lambda_1 = 1.89$ and $\lambda_2 = 0.78$, so that the resulting surface response is a minimum response [9]. The response surface model can be seen in the following figure:

![Figure 1. Response surface curve % bioplastic residue.](image-url)

Based on the surface response curve of the residual % in Figure 1 above, it can be seen that the response characteristics obtained in this study are in the form of a minimum response. The higher the concentration of glycerol used, the higher the biodegradation value of the bioplastic, resulting in a lower % residual. The use of bioplastics from bio-based materials, namely starch which contains cellulose has a hydroxyl group OH- which is able to absorb water contained in the soil, so it really helps accelerate the decomposition process by soil microorganisms, so that bioplastics will be eroded and turned into much smaller pieces caused by because of the breaking of the polymer chain bonds, so that in the end the pieces of bioplastic will disappear over time [10].

3.2. Analysis response water resistance

Based on the results of data processing, the equation for the % water resistance is in the form of a quadratic which can be written as follows:

$$Y = +81.45358 + 4.82637X_1 - 2.41997X_2 - 0.079917X_1X_2 - 0.36116X_1^2 - 0.088411X_2^2$$  \hspace{1cm} (2)$$

Based on the data in Table 2, it can be seen that the variables that affect the % water resistance response are $X_1$, $X_2$, and $X_1^2$. From equation (2), the eigenvalues of the matrix are both negative namely $\lambda_1 = -3.45$ and $\lambda_2 = -0.603$, so that the resulting surface response is a maximum response. The response surface model can be seen in the following figure:
Based on the surface response curve of the % water resistance in Figure 2 above, it can be seen that the response characteristics obtained in this study are in the form of a maximum response. It can be seen that the absorption of bioplastics to water is higher with increasing glycerol concentration, as well as the effect of ZnO where the higher the concentration of ZnO, the smaller the absorption of bioplastics to water. The hygroscopic nature of glycerol and having an OH\(^{-}\) group is able to bind to H\(_2\)O through hydrogen interactions, this is what affects the absorption of bioplastics to water higher with increasing glycerol concentration [11], thus causing lower water resistance.

3.3. Analysis response heat resistance

Based on the results of data processing, the equation for the % head resistance is in the form of a quadratic which can be written as follows:

\[
Y = +59.11694 + 8.94138X_1 - 0.16207X_2 + 0.23611X_1X_2 - 0.55208X_1^2 - 0.24653X_2^2
\]  

(3)

Based on the data in Table 2, it can be seen that the variables that affect the % head resistance response are \(X_1\), \(X_2\), and 2FI (the interaction of two factors). From equation (3), the eigenvalues of the matrix are both negative namely \(\lambda_1 = -6.12\) and \(\lambda_2 = -1.059\), so that the resulting surface response is a maximum response. The response surface model can be seen in the following figure:
Based on the surface response curve of the % water resistance in Figure 3 above, it can be seen that the response characteristics obtained in this study are in the form of a maximum response. It can be seen that the higher the concentration of ZnO used, the higher the heat resistance value (°C) of the bioplastic. Likewise with the concentration of glycerol, where the higher the concentration of glycerol used, the lower the value of heat resistance (°C) of the bioplastic.

3.4. Value of Response Optimization of Physical Characteristic of Bioplastic

The optimal condition of the response given can be seen in Table 3 below:

| Name                | Goal        | Lower Limit | Upper Limit |
|---------------------|-------------|-------------|-------------|
| ZnO (%)             | is in range | 1.76        | 10.24       |
| Glycerol (ml)       | is in range | 0.76        | 9.24        |
| Residual (%)        | Minimum     | 75.979      | 92.176      |
| Water resistance (%)| Maximize    | 70.333      | 96.976      |
| Heat resistance (%) | Maximize    | 68          | 98          |

Source: Data processing results

Based on the results of the response optimization presented in Table 3 above, the optimization graph is obtained which is presented in Figure 4 and 5 below:

**Figure 4.** Optimum response surface curve for bioplastic physical properties (contour)
Based on Figures 4 (contour) and Figure 5 (3D surface), the recommended solution for the ZnO variable is 7.42% and the glycerol variable is 4.96 mL with optimal response conditions for the % residual of 81.3612%, water resistance of 86.1333%, and heat resistance of 96.888°C. From the solution given, the resulting desirability value is 0.725 which indicates that the formula will produce optimal and desirable physical characteristics of bioplastics of 72.5%.

4. Conclusion

The combination of the basic ingredients of agricultural waste in the form of jackfruit seeds and corn cobs is able to produce bioplastics with good physical properties, where the optimization of the response to the physical properties of bioplastics recommended by the program is for the ZnO variable of 7.42% and the glycerol variable of 4.96 mL with the optimum value residual is 81.3612%, water resistance is 86.1333%, and heat resistance is 96.888°C with a desirability value of 0.725. The use of concentrations of ZnO and glycerol affects the physical properties of the resulting bioplastic, where the higher the use of ZnO, the water resistance and heat resistance of the bioplastic will increase, while the higher the concentration of glycerol used, the value of % residual will be lower.

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