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Additional Nata de Coco on The Performance of Cassava Starch Based Bioplastic

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Abstract

Bioplastics, an environmental friendly materials, attracting interest to many scientists around the world. Cassava starch may used as a major ingredient for bioplastics production. The addition of cellulose could improve the quality of starch base bioplastics. Cellulose used for thispurpise is mainly extracted from wood. Extraction of cellulose from wood materials is not easy. Removing contaminant particularly lignin is the major obstacle. The use nata de coco, a pure crystalline cellulose, may of great choice to replace cellulose of wood origin. It is the aim of this study to search the possibility of using nata de coco as filler replacing cellulose of wood origin for the production of bioplastic using cassava starch. The effect of nata de coco as a filler on the quality of bioplastics was studied using a Central Composite Design (CCD) and Response Surface Analysis (RSA). The ratio of tapioca and nata de coco and the ratio of nata de coco and glycerol were evaluated. The performance of bioplastics was measured semi qualitatively based on the structure, color, and its similarity to synthetic plastics. The result shows that the quality of bioplastics affected by nata de coco linearly when the maximum of nata de coco used up to 40% (v / v). Bioplastics may be prepared using 10% (w / v) tapioca, 3% (w / v) glycerol and 40% (v / v) nata de coco. The average degree of swelling of bioplastics is 18.75%, an average tensile test of 0.01494 kgf / mm^2 and an average elongation 14.4056%. The results of this study indicate that nata de coco may be used as a filler in the manufacture of starch-based bioplastics re placing wood based cellulose.

Keywords: Biodegradable, Bioplastic, Nata de coco, Plastic

INTRODUCTION

Plastic is a chemical polymer used in food packaging. Polyethylene (PE), polypropylene (PP), polystyrene foam (PS) and polyethylene terephthalate (PET) are the most common plastics used for this purpose. The advantages of plastics are inexpensive, easy to manufacture, excellent mechanical performance, excellent protective properties on the packaged products.

Plastic, however, is very difficult to degrade naturally, causing environmental pollution. Over time, plastics may also be fragmented in nature into small pieces to form microplastics, measuring 0.1 μm - 5 μ. In this form plastic particles not only pollutes environment but may also enter the food chain which eventually affecting living things, including human health. Microplastics can also bind to heavy metals, which has led to even more serious problems. Addressing plastic pollution, including plastic pollution in the sea, will certainly take a very long time, expensive, and inefficient (Beaumont, 2019).

It is obvious, that environmental pollution by plastics steady increased worldwide. This situation, attracting scientists around the world to develop alternative materials to plastics which is more environmentally friendly and more sustainable. Much attention has been focused on developing biomass as raw material for the production of bioplastic, much easier to degrade (biodegradable) compared to petroleum based plastics (Muthuraj, 2016). Microcrystalline cellulose used as reinforcement filler for starch based edible films was analyzed. The results showed higher strength and elongation and lower Water Vapour Transmission rate (WVTR) of bioplastics (Maulida, 2016). The derivation of high-quality wood pulp that produces crystalline cellulose can be fragmented into cellulose fractions as a hydrolysis product. The diameter of MCC can be around 100 μm (Abdullah et al, 2020). However, there are more smaller type of crystalline called Nanocrystalline Cellulose. The diameter of Nanocrystalline Cellulose (NCC) typically varies from 5 to 20 nm and the length of the particles is observed to be between 100 nm to several micrometers (Purkait *et al*, 2021). Therefore, Nanocrystalline has a higher specific surface area than other cellulose fibers which is good for improving the mechanical properties of starch bioplastics.

Agustin *et al*. (2014) successfully produced bioplastic from starch and find that cellulose nanocrystals is essential as matrix to improve bioplastic performance. Meneguina *et al*. (2017) also use nanocellulose as a filler to produce a good quality bioplastics. Agustin *et al*, (2014) preparing cellulose nanocrystal from rice straw. It needs special effort to purified rice straw cellulose from its impurity such as lignin. To produce cellulose nanofiber from rice straw requires complicated processes as the cellulose present as a hemicellulose or strongly bound to lignin (Hervy, 2018). For this reason, there is a need to find a new in expensive and readly available cellulose material. Nata de coco is one of the choices. Nata de coco is a pure crystalline cellulose produced by *Acetobacter xylinum* through fermentation of sugary rich material e.g. coconut water. Nata de coco may possibly used directly as a matrix for the production of bioplastics from starch without treatment. The sue of nata de coco for the production of bioplastic from cassava starch (tapioca) has not been reported so far. The aim of this study is to find out the effect of nata de coco pulp as matrix to improve the performance of tapioca starch bioplastic.

MATERIAL AND METHODS

Materials

Cassava is obtained from the market. A total of 2 kg of cassava is peeled and cleaned, cut into small pieces. A 2 L of clean water is added then mashed in a blender and filtered through a cheese cloth. The starch suspension is taken and the remaining starch in solid particles is again extracted with the addition of more water repeatedly then filtered with cheese to obtain starch suspension. Starch suspensions were left for 24 hours to obtain starch sediment. The water on the top part is removed by decantation and the starch sediment collected. The starch then dried using oven at 40 - 50 ^{o}C. The starch is then ground and sifted through a 140 stainless steel wire mesh to produce fine tapioca flour. Nata de coco sheets are purchased online and then processed into nata de coco pulp.

Methods

To optimized the ratio of tapioca starch and nata de coco pulp, the experiment was designed using the Central Composite Design with 2 variables and 5 levels of combination (table 1 and 2). The same experiment was also designed similarly to optimized nata de coco pulp and plastisizer, gliserol. The parameters observed include texture, color, and its comparation to synthetic plastics. The observations were converted to a semiquantitative value between 1 and 5 indicating the quality of bioplastic (very bad, bad, nomal, good, and very good).

Table 1. Ratio of tapioca and nata de coco pulp on the performance of biopastic

No.	Tapioca suspension (mL)	Nata de coco suspension (mL)
1	1.58579	5
2	2	10
3	2	10
4	3	5
5	3	10
6	3	15
7	3	2.92893
8	3	10
9	3	17.0711
10	3	10
11	4	10
12	4	10
13	4.41421	15

Table 2. The ratio of glycerol and nata de coco pulp on the performance of bioplastic

No.	Glycerol Suspension (g)	Nata de coco suspension (mL)
1	0,585786	25
2	1	10
3	1	40
4	2	46,2132
5	2	25
6	2	25
7	2	3,7860
8	2	25
9	2	25
10	2	25
11	3	10
12	3	40
13	3,41421	25

Another experiment conducted to observe the addition of citric acid on the performance of tapioca based bioplastic.

Bioplastic Preparation

A total of 10 g of tapioca was dissolved in 200 mL of distilled water and heated in a water bath at a temperature of 60° C for 15 minutes to obtain tapioca suspension. Main while, to obtain nata de coco pulp, a total of 100 g of wet nata de coco sheets are cut into cubes (approximately 1 cm^3 in size), mixed with 100 mL of distilled water and blended for one minute at maximum speed until it becomes fine nata de coco pulp. Tapioca suspension and nata de coco pulp were mixed according to the CCD design. To the mixture of tapioca suspension and nata de coco pulp, the amount of 6 g sorbitol and 3 g glycerol are added and stirred for two hours at 70°C. Bioplastics is prepared by pouring the mixture into a glass mold that was previously cleaned with 96% alcohol and coated with vegetable oil. The material is then heated in an oven at 60 ° C for 24 hours. The bioplastic were removed from mold, cooled at room temperature and observed.

Swelling Test

The Swelling test is carried out by adding a 5 drops of water along the surface of bioplastic tested. After 2 minutes, the water is drained and the remaining water is absorbed using a tissue and weighed. The degree of swelling is calculated using the formula:

$$S = \frac{m2 - m1}{m1} x\ 100\%$$

S = Degree of swelling
m1 = The final mass of the sample
m2 = The initial mass of the sample

Measurement of Tensile Strength and Percentage of Elongation (ASTM E252)

Tensile strength measurements were carried out on bioplastic samples. For this purpose the bioplastic is cut to a size of 10 x 2.5 cm. Both ends of the bioplastic are clamped and the initial length is measured. After that a load is added to the bioplastic that has been clamped until the bioplastic is cut off, and the final length of the bioplastic is recorded when it is cut. The tensile test strength is calculated using the formula:

$$\sigma = \frac{F}{A}$$

σ = Tensile strength
F = Added load
A = Bioplastic surface area

Calculating the elongation value:

$$\varepsilon = \frac{\Delta 1}{10} x\ 100\%$$

E = Bioplastic elongation value
Δl = Difference between the initial length of the bioplastic and the length of the end of the bioplastic
l0 = Bioplastic starting length

Data analysis

The effect of tapioca suspension and nata de coco pulp and the effect of glycerol and nata de coco pulp obtained from the experiment designed using CCD, were analyzed using Response Surface Analysis (RSA) by the aid od statistical program Design Expert 11.

RESULT

Bioplastic Quality

The effect of tapioca suspension and nata de coco pulp is shown in Table 3.

Table 3. Response ratio of tapioca and nata de coco to quality of bioplastics

No.	Tapioca suspension (mL)	Nata de coco suspension (mL)	Result		
			Texture	Warna	Bioplastic quality (Conversion of qualitative values to quantitative values)
1	1.58579	5	A little sticky	Clear white	3
2	2	10	Not sticky	Clear white	4
3	2	10	Not sticky	Clear white	5
4	3	5	Sticky	Clear white	2
5	3	10	A little sticky	Clear white	3
6	3	15	A little sticky	Clear white	4
7	3	2.92893	Sticky	Clear white	1
8	3	10	Not sticky	Clear white	4
9	3	17.0711	Not sticky	Clear white	5
10	3	10	Not sticky	Clear white	4
11	4	10	Not sticky	Clear white	5
12	4	10	A little sticky	Clear white	4
13	4.41421	15	Not sticky	Clear white	5

Information: 1 = very bad, 2 = bad, 3 = good, 4 = very good, 5 = excellent

The results of ANOVA analysis, the concentration of tapioca suspension and nata de coco pulp at the range tested were not significantly affecting the quality of bioplastic. With more careful observation, the effect of nata de coco pulp, however is significant linearly with a p-value of 0.0006.

Meanwhile, the effect of glycerol and nata de coco pulp ratios is illustrated in table 4. The results of ANOVA analysis show that the effect of nata de coco pulp is also significant linearly with a p-value of 0.0001.

Table 4. Response to the ratio of nata de coco and glycerol to the quality of bioplastics

No.	Glycerol (g)	Nata (mL)	Hasil		
			Texture	Warna	Bioplastic quality (Conversion of qualitative values to quantitative values)
1.	0,585786	25	Not sticky	Clear white	4
2.	1	10	Sticky	Clear white	2
3.	1	40	Not sticky	Clear white	4
4.	2	46,2132	Not sticky	Clear white	5
5.	2	25	Not sticky	Clear white	4
6.	2	25	A little sticky	Clear white	3
7.	2	3,7860	Sticky	Clear white	1
8.	2	25	A little sticky	Clear white	3
9.	2	25	Not sticky	Clear white	4
10.	2	25	Not sticky	Clear white	4
11.	3	10	Sticky	Clear white	2
12.	3	40	Not sticky	Clear white	5
13.	3,41421	25	Not sticky	Clear white	4

Information: 1 = very bad, 2 = bad, 3 = rather good, 4 = good, 5 = very good

Based on the above results, bioplastic sample was again prepared using a mixture of 10 g of tapioca, 3 g of glycerol, 6 g of sorbitol and 40 mL of nata de coco pulp per 100 mL of bioplastic dough. With this composition, a bioplastic is also prepared using 100, 200 and 300 mL of the same mold capacity to produce bioplastics with different thicknesses. Using the same method, a bioplastic was also made by adding 0%, 0.5%, and 1% (v / v) citric acid. The addition of citric acid is intended to improve the hydrophobic properties of the bioplastic. Bioplastic produced were tested for their swelling, tensile strength, and elongation properties.

The inflation test is carried out to see how much water being absorbed by the bioplastic. The higher the water content absorbed, the worse the quality of the bioplastic. The results of the inflation test are shown in Table 5.

Table 5. Effect of bioplastic thickness on the degree of swelling

No.	**Types of Bioplastics**	**Initial weight (g)**	**Weight after adding water (g)**	**Result (%)**
1.	A	8	9	12,5%
2.	B	8	10	25%
3.	C	9	11	25%

Information: Bioplastics are made with the same mold size, A = made using a volume of dough (a mixture of tapioca, nata de coco pulp, glycerol and sorbitol) 100 mL, B = using a dough with a volume of 200 mL and C = using a dough with a volume of 300 mL.

The swelling test show different results. Bioplastic A had a degree of swelling of 12.5%, while bioplastic B and C had a degree of swelling of 25%. Citric acid added to compare the hydrophobic of the bioplastic. The results are shown in Table 6.

Table 6. Effect of addition of citric acid on the degree of swelling of the bioplastic

No.	Types of Bioplastics	Before weighing it (g)	After weighing it (g)	Hasil (%)
1.	A	8	9	12,5%
2.	B	8	9	12,5%
3.	C	8	10	25%

Information: Bioplastics are made with the same mold size, A = made using a volume of dough (a mixture of tapioca, nata de coco pulp, glycerol and sorbitol) 100 mL, B = using a dough with a volume of 200 mL and C = using a dough with a volume of 300 mL.

The swelling test on the bioplastic sheet made with the addition of citric acid able to limit the amount of water absorbed to 12.5%.

The tensile and elongation tests were carried out using a Shimadzu Autograph machine using the American Society for Testing and Materials (ASTM) E252. The sample to be tested is cut to a size of 10 x 2.5 cm. Tensile tests were carried out on bioplastics of different thicknesses. The results are Table 7.

The tensile test results show that thicker bioplastics have the highest tensile test, 0.01845 kgf / mm2, but the shortest elongation, 13.8376%. Meanwhile, the results of tensile and elongation tests for bioplastics made with the addition of citric acid are illustrated in Table 8.

Table 7.The results of tensile and elongation tests for bioplastic products using the Shimadzu Autograph machine with the American Society for Testing and Materials (ASTM) E252 (ASTM E252) method.

No.	Types of Bioplastics	Tensile strength (kgf/mm^2)	Elongation (%)
1.	A	0.01263	14.6243
2.	B	0.01375	14.7550
3.	C	0.01845	13.8376

Information: Bioplastics are made with the same mold size, A = made using a volume of dough (a mixture of tapioca, nata de coco pulp, glycerol and sorbitol) 100 mL, B = using the volume of dough (a mixture of tapioca, nata de coco pulp, glycerol and sorbitol) 200 mL and C = using dough (a mixture of tapioca, nata de coco pulp, glycerol and sorbitol) with a volume of 300 mL. The whole dough is poured into the molds of the same size.

Table 8.The results of tensile and elongation tests for bioplastic products with the addition of citric acid using a Shimadzu Autograph machine using the American Society for Testing and Materials (ASTM) E252 (ASTM E252) method.

No.	Types of Bioplastics	Tensile strength (kgf/mm^2)	Elongation (%)
1.	A	0.00388	15.4578
2.	B	0.00405	16.4637
3.	C	0.00549	11.3778

Information: A = 100 mL of dough (a mixture of tapioca, nata de coco pulp, glycerol and sorbitol) is added with 0% (w / v) citric acid, B = 100 mL of dough plus 0.5% (w / v) citric acid, and C = 100 mL of dough plus 1% (w / v) citric acid.

The highest tensile value is shown by the thickest bioplastic with a figure of 0.00549 kgf / mm2.

DISCUSSION

The concentration of nata de coco as matrix/filler for bioplastic need to be increased much higher. The use of nata de coco in the form of pulp, should be very difficult. For this reason it is suggested that nanofiber material should be prepared in a powder form before it is used as a matrix in the production of bioplastic. Meanwhile, the effect of glycerol, both in linear and quadratic region do not significant on the quality of bioplastic produced. For this reason the concentration of biocellulose need to be increased. Again the preparation of nanofiber powder from nata de coco is required.

The degree of swelling can be interpreted as the amount of water absorbed by bioplastic which indicate its hydrophilic properties. This property is related to the presence of glycerol and tapioca. With the increasing concentration of glycerol and starch, the ability of bioplastics to absorb water is higher (Basiak, 2018). This test is very important to determine whether or not bioplastic can withstand exposure to water. The greater the water absorption, the lower its ability to withstand water exposure, in other words, the product will deteriorate more quickly or reduce its quality if exposed to water (Yanti, 2017). From the results of this test, thinner bioplastics are better. Maulida *et al* (2016) research result from Microcrystalline Cellulose Avicel PH 101 can take around 65- 75% water. Meanwhile

Nanocrystalline Cellulose from Nata de coco can take water from 12,5- 25 %. It still advantage using Nanocrystalline Cellulose Nata de coco as reinforce plasticizer. The hydrophobic properties still need to be improved. For this reason, citric acid is added. The reaction between carboxyl groups in citric acid and hydroxyl groups in starch is expected to form strong bonds and can reduce free hydroxyl groups in starch which will make starch more hydrophobic (Kawijia, 2017). The addition of citric acid, however was not able to significantly improve the quality of the bioplastic. This result also occurred in Marichelvam's (2019) study, where water absorption was in the range of 11.7 and 13.9%.

Thickness can increase the tensile but decrease the elongation property. The mechanical properties of bioplastic films are influenced by the large number of constituent components that cause the thickness of the bioplastic (Santana et al., 2018). The addition of citric acid as illustrated in Table 8, was not able to significantly improve the tensile strength of the bioplastic. When compared with the tensile value of a bioplastic of the same thickness but not added with citric acid, the tensile strength is much lower. The tensile strength of bioplastics without citric acid is 0.01845 or 29.75 times stronger. On the other hand, the elongation performance tends to be better than that of bioplastics without citric acid mixtures. The best elongation performance was 16.4637% with the addition of 0.5% citric acid better than the same bioplastic made without the addition of citric acid. However, the addition of citric acid at higher concentration (1.0%), the elongation strength reduce to 11.3378% or lower than the same bioplastic made without citric acid which was 18.8376%. To a certain extent, the addition of citric acid will reduce the interaction between starch molecules and can act as a plasticizer (Wilpiszewska, 2019). The Indonesian State Standard (SNI) 7818: 2014 regarding biodegradable plastic bags has a tensile strength standard of 1.37 kgf / mm2 and an elongation of 400 - 1220%. The results of the tensile and elongation tests achieved in this experiment are still far from the SNI standards. The addition of glycerol and sorbitol which play a role in the process of increasing the plasticity of bioplastics while the addition of nanofiber as a matrix to improve the tensil strength of bioplastic needs to be further investigated to obtain bioplastics that have soft, ductile, and strong mechanical properties (Suryanto, 2016). Jannah's research (2019) states that the elongation properties are influenced by cellulose fibers as bioplastic fillers, although they are only able to increase the elongation properties by 4.75%. Maulida research (2016) and Abdullah research (2020) using MCC resulting under 10% elongation. It still advantage using Nata de coco Nanocrystalline Cellulose as a reinforce bioplastic. But, Increasing the concentration of nata de coco fibers needs to be investigated further to obtain bioplastics with high elongation power and tensile strength closer to the SNI standard.

CONCLUSION

It is concluded that Nata de coco can be used as a filler in the manufacture of bioplastics using tapioca as the main raw material. The addition of nata de coco pulp significant effecting bioplastics properties linearly. Bioplastics can be made using 10% (w / v) tapioca, 40% (v / v) nata de coco pulp, 3% (w / w) glycerol and 6% (w / w) sorbitol. The addition of citric acid can affect the hydrophobicity, degree of swelling and elongation but reduce the tensile strength. To produce bioplastic that met the SNI standard, the concentration of biocellulose fiber to tapioca with the right concentration of glycerol is required. It is also the challenge to search and identified a proper application of bioplastic produced in this experiment but may need to develop new SNIs for the bioplastic produced in this study, including for food coating or capsule shells in the pharmaceutical industry.

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