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Synthetic Bioplastics from Cassava Skin using the Nata Method and the Addition of Plasticizers

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Abstract. Plastics have an important role in human life. But the difficulty of plastic degradation makes the emergence of much research to produce plastics that are easily degraded. Bio-based plastics are a major attraction for addressing environmental challenges and sustainable issues. The purpose of this study is to make bioplastics from cassava skin by fermentation. Cassava skin is a common waste found in Indonesia which is just thrown away so it can pollute the environment. Cassava skin contains starch which is the main material of making biodegradable polymer. In this study, the plastics were synthesized using nata method, where the content of cassava starch was fermented with *Acetobacter xylinum* bacteria and given plasticizer addition. Variation of plasticizer type (1.4 butanediol and polyethylene glycol) and variations of its concentration (0.5%, 0.1%) were done this study. The addition of a plasticizer is intended to improve the quality of the mechanical properties of the synthesized film. The results were in the form of bioplastic sheet and tested for characteristics in accordance with the mechanical properties from bioplastic which are among others of tensile strength, elongation and biodegradability. The best result is bioplastic sheet with the addition of 0.5% polyethylene glycol plasticizers have better mechanical properties with a tensile test value of 312.93 MPa (31.91 kgf/mm), an extension of 5.09% and the rate of degradation in the soil 0,02gram / day. FTIR spectrum shows that the main chemical groups are OH, N-H (3342.03 cm^{-1}), C-O (1054.87 cm^{-1}), C = C (1646.91 cm^{-1}) and NO₂ (1316.21 cm^{-1}). This research successfully produced bioplastic sheet by Nata method using 0,5% polyethylene glycol as plasticizer and cellulose from fermentation skin cassava starch as matrix. In the future prospects, this bioplastic sheet can be developed as plastic bags and food packaging.

INTRODUCTION

Bioplastic is a type of polymer composed of biomass that can produce or commonly called biopolymer. Biopolymer can be degraded easily, until bioplastics are more environmentally friendly than synthetic plastics. The use of biomass as a base for new bioplastics. The history of bioplastics can be traced back to 1900 when Henry Ford businessman developed a method for making bioplastic from soybean to be used in car [1]. Therefore, biodegradable polymers can make significant contributions to material recovery, reduction of landfill and utilization of renewable resources [2].

One solution for natural ingredient that can be used as a base for making bioplastic is starch [3]. Starch is determined by the α -glycosidate chain. The compound, which only produces hydrolysis, is a homopolymer called glucosan or glucan. Two main elements are amylose (15-20%) which has a branchless helical structure, and amylopectin (80-85%) which consists of several branched chains composed of 24-30 glucose residues which at the branch point are joined by bond 1 \rightarrow 4 and 1 \rightarrow 6 in the chain. Glucose is formed through photosynthesis and for energy or can be used as starch which will later [4].

Indonesia is rich in plants that contain starch [3]. For example, cassava, which is the food of the Indonesian people [5]. But if we process cassava as a material for making polymers, we will disturb the condition of food security in the country. Thus, it is necessary to do alternatives other than cassava. This research tried to use the wasted cassava parts, namely the cassava skin [3].

Cassava skin is peeled waste produced by chips, tapioca, cassava, tape, and other cassava-based snacks or food in several regions in Indonesia. The potential of cassava skin in Indonesia is very abundant, along with the existence of this country as one of the largest cassava producer in the world and increasing every year [5].

Cassava peels contain 44-59% starch, which is a good condition for bacterial growth of *Acetobacter xylinum* in the manufacture of Nata [1]. Nata is the result of bacterial fermentation of *Acetobacter xylinum* which forms gel sheets on the surface of the substrate in the form of cellulose [1]. *Acetobacter xylinum* belongs to the Pseudomonas family and belongs to the genus Acetobacter. , The specific nature of this bacterium is its ability to form a thick membrane on the surface of the fermentation medium called cellulose [6]. Biodegradable plastics, based on cellulose acetate, were studied and the produced plastic decomposed in soil or water within a few years. However, the material can be recycled, also, or incinerated without residue [7].

Cellulose has a potential to be used as a source for bioplastic production. Usage of cellulose in bioplastic production need some modification. Derivate of the cellulose that has been used in bioplastic synthesis was cellulose nano-crystals (CNC) [8], nano-fiber cellulose (NFC) [9], cellulose acetate butyrate [10], cellulose acetate [11] and bio-PE [12]. Cellulose is a β -1,4 poly glucose, with a very large molecular weight. Repeat units of cellulose polymers are bound through glycoside bonds which result in linear cellulose structure. The regularity of the structure also causes hydrogen bonds intra- and intermolecularly. Some cellulose molecules will form microfibrils with a diameter of 2-20 nm and a length of 100-40000 nm, some of which are regular (crystalline) regions and interspersed with an amorphous region that is irregular. Some microfibrils form fibrils which eventually become cellulose fibers. Cellulose has a high tensile strength and is insoluble in most solvents. This is related to fiber structure and the strength of hydrogen bonds [13].

In this study, the process of bioplastic processing of cassava peel was carried out in 5 stages, namely flour making, material mixing, polymer formation with nata method by fermentation using the bacterium *Acetobacter xylinum*, and the process of plastic printing.

In the processing of bioplastics plasticizers are needed, in this study comparing bioplastic fermentation with the addition of Plasticizer. The addition of plasticizers both synthetic and natural is believed to improve the properties of the bioplastics produced, expand or modify their basic properties or can create new properties that are not present in the basic ingredients [14]. The plasticizers used in this study were polyethylene glycol (PEG) and 1,4-butanadio[15].

Polyethylene glycol or with the name IUPEC Alpha-Hydro-Omega-Hydroxypoly (oxy-1,2-ethanadiol) are compounds with chemical formula $(C_2H_4O)_n + H_2O$ and structure formula $HOCH_2-(CH_2-O-CH_2)_n-CH_2OH$. Polyethylene Glycol is a long chain polymer compound, not inert with a molecular weight between 200-9500 Da. Whereas 1,4-Butanediol, colloquially known as BD, is an organic compound with the formula $HOCH_2CH_2CH_2CH_2OH$. This is a colored thick liquid. This is 1-4 butanediol stable isomers [16].

MATERIAL AND METHODS

Materials

The materials used in this study are: (1) Cassava skin waste collected from markets and fried vendors on the street between Babelan- Jl. Perjuangan, Bekasi, West java, Indonesia, (2) Glacial acetic acid, (3) Aquadest, (4) Sugar, (5) Urea, (6) Acetobacter Xylinum (Agrotekno, Bogor, Indonesia), (7) 1,4-Butanediol (BD Merck), and (8) Polyethylene Glycol (PEG) 40 (Merck).

Methods

Nata Making Stage [1]

Before making bioplastics, organic materials are first converted into nata. In this case, the experiment used cassava skin starch. The starch was then boiled. At the time of boiling, 2.5% of sugar and 0.5% urea of the amount solution were added. After boiling, filtering was done to get the juice from the boiling ingredients. The filtered solution was then cooled. After cold, the addition of Acetobacter xylinum and acetic acid was done to maintain the pH in the range of 3.0–4.0. Furthermore, to determine the effect of adding plasticizers to the mechanical properties of bioplastics, solution of 1,4-butanediol and PEG 40 was added. Then fermentation treatment was done for 5 days.

Drying Phase

Drying phase of nata includes: (a) pressing into a film form and (b) drying nata for two days becomes a film by air and not exposed to direct sunlight.

Characteristics (shimadzu autograph-based on ASTM D638)[17]

1. Tensile Strength

The samples to be tested are first conditioned in a room with standard temperature and humidity ($23 \pm 20\text{C}$, 52%) for 24 hours using Tensile Strength. The sample to be tested was cut to a standard of 2 x 8 cm. The test was carried out by means of both ends of the sample clamped. Then the initial length was recorded before the addition of the load. After recording the clamped film is added to the load. Then the next sheet was tested using Equation (1).

$$\text{Tensile Strength } \left(\frac{\text{Kgf}}{\text{mm}^2} \right) = \frac{\text{Tensile Force (F)}}{\text{Surface Area (A)}} \quad (1)$$

2. Elongation

Elongation measurements were carried out in the same way as tensile strength testing. Elongation is expressed as a percentage, as shown in Equation (2).

$$\text{Elongation (\%)} = \frac{l-l_0}{l_0} \times 100\% \quad (2)$$

Description: I = length after breaking up
I₀ = length at first

3. Biodegradation Test [18]

Biodegradation test is used to determine the degradation ability of biodegradable plastic samples by placing bioplastic samples in soil containing manure. After ten days, the sample was taken, cleaned then weighed. In the biodegradation process, the standard test that can be carried out is the mass loss test and the rate of mass loss in a certain period of time. This value can be calculated using Equation (3).

$$\text{Mass loss (\%)} = \frac{w_i - w_f}{w_i} \times 100\% \quad (3)$$

Where:

W_i = mass of the sample before incubation.

W_f = mass of the sample after biodegradation.

The determination of the mass loss rate (v) can be calculated using Equation (4).

$$v = \frac{w_i - w_f}{\Delta t} \quad (4)$$

Where:

v = the rate of mass loss.

t = time needed for biodegradation

Fourier Transform Infrared (FT-IR) analysis (Shimadzu IRPretige-21) [19]

FTIR analysis was carried out to determine the functional groups contained in bioplastics from fermented cassava skin starch with the addition of plasticizers. Data analysis was performed by observing the absorptions that appeared on each FT-IR spectrum.

RESULTS AND DISCUSSION

One of the parameters that can determine the quality of biodegradation plastic film are the mechanical properties of plastic films, consisting of tensile strength, percent elongation-to-break and elasticity (elastic / young modulus) [20]. In this study two characteristic tests were carried out namely tensile strength and elongation strength.

In this study, cellulose from starch fermentation by the bacterium *Acetobacter xylinum* in the form of nata was used. Nata is mucus released by *Acetobacter xylinum* which is the result of the secretion of sugar metabolism. Both starch content and added sucrose will be hydrolyzed to glucose and converted by bacteria through a biochemical process into cellulose. Cellulose formed is threads together with slimy polysaccharides form a continuous link continues to be a layer of nata. CO₂ bubbles produced during fermentation has a tendency to attach to this layer, thus causing the layer lifted to the surface of the liquid [1]. The layer taken as cellulose which will be mixed with plasticizer forms bioplastic [21]. Figure 1 shows the relationship between the addition of plasticizers and tensile strength of bioplastic films.

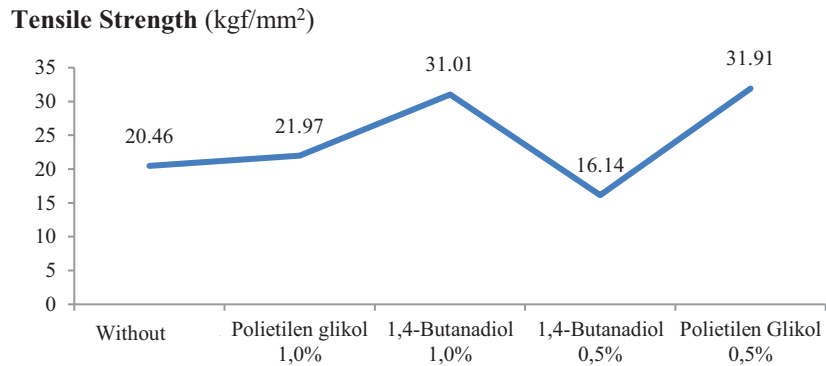


FIGURE 1. The effect of treatment on tensile strength

It can be seen in Figure 1 that the biggest tensile strength is biodegradable plastic with the addition of plasticizer 0.5% polyethylene glycol with an average tensile strength of 312.93 MPa (31.91 kgf / mm²). While the lowest average tensile strength is biodegradable plastic with the addition of 0.5% butanadiol plasticizer with an average tensile strength of 158.27 MPa (16.14 kgf / mm²).

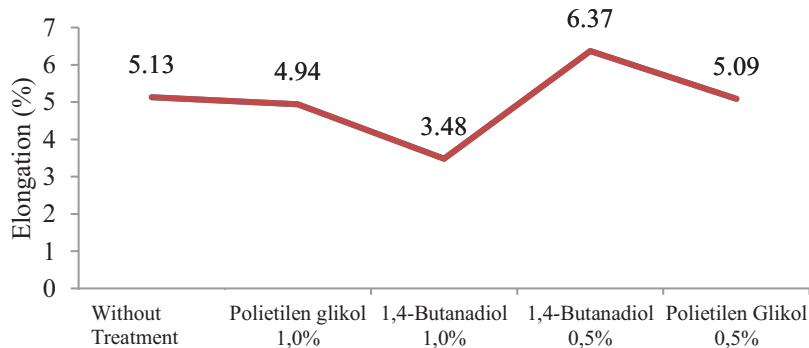


FIGURE 2. The relationship of the addition of plasticizers with elongation-to-break

Figure 2 shows the relationship of adding plasticizers with elongation-to-break. From Figure 2 it can be seen that the highest elongation at the addition of 1,4-Butanadiol 0,5% and on the addition of plasticizers 0.5% polyethylene glycol has a value of 5.09%. Based on the nature of the polymer, the higher the tensile strength, the greater the modulus of elasticity, while the extension becomes smaller. Thus, the tensile strength has a large influence on the mechanical properties of a polymeric material [9].

Figure 3 shows the results of the biodegradation test. Biodegradation test is a requirement for determining the quality of bioplastic which is declared as a plastic that is easily degraded in nature.

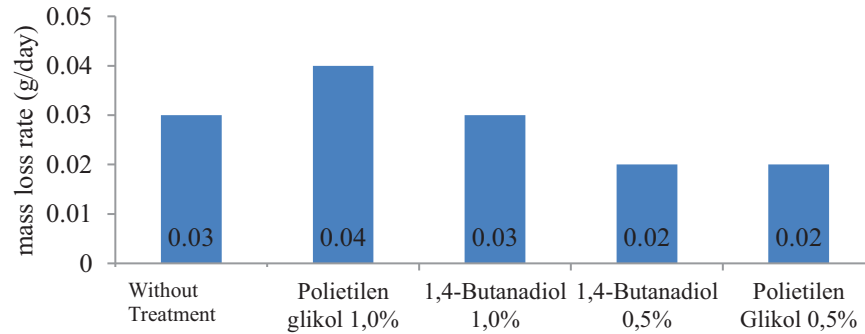


FIGURE 3. The rate of biodegradation test results

From figure 3, it can be seen that the highest mass loss rate is biodegradable plastic with the addition of plasticizer 1% polyethylene glycol with a mass loss rate of 0.04 g / day. While the addition of 0.5% polyethylene glycol the rate of mass loss is 0.02 g / day. This mass loss rate is influenced by the ease of the biodegradable plastic film material to be decomposed by microorganisms. Based on ASTM D 5988 [22], the material is said to be biodegradable if it can be degraded at least 90% after 6 (six) months.

Characterization of bioplastic functional groups from cassava peel was determined by Fourier Transform Infra-Red Spectrophotometer (FTIR) and the results of the monitoring of functional groups are presented in Figures 4,5, and 6 below [23] and [24].

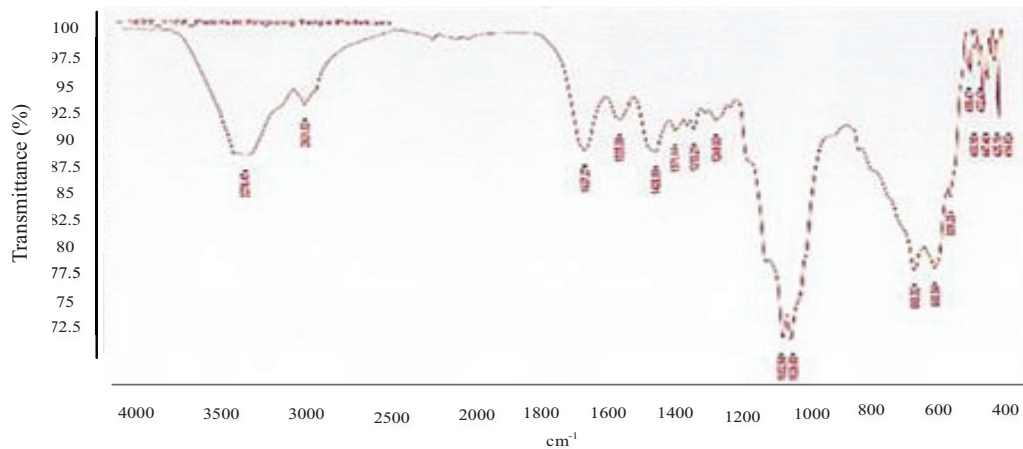


FIGURE 4. Bioplastic spectrum without treatment

Figure 4 identified O-H carboxylic acid group at the peak point 3276,47 cm^{-1} , alkene group C=C at the peak point of 1637,27 cm^{-1} , C-O group at the peak point of 1029,50 cm^{-1} .

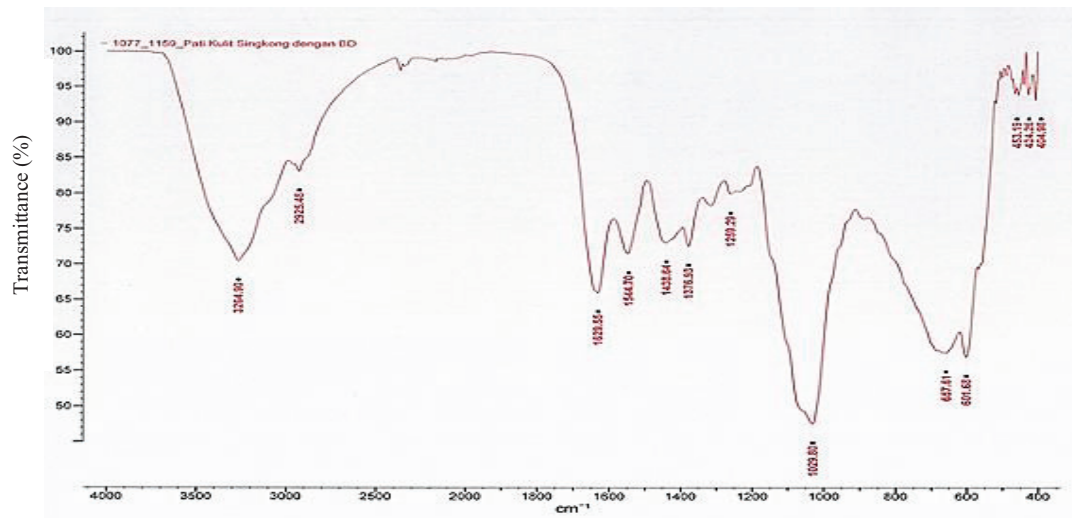


FIGURE 5. Bioplastic spectra with the additional of 1,4-Butanediol

Figure 5 identified ether C-H and carboxylic acid O-H group at the peak point 3264.90 cm^{-1} , alkene and aromatic group, C=C at the peak 1629.55 cm^{-1} .

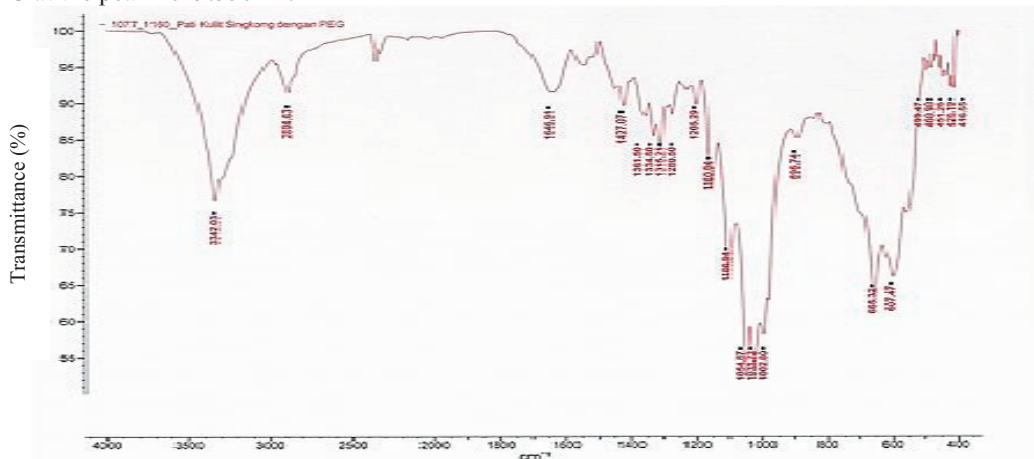


FIGURE 6. Bioplastic spectra with the addition of polyethylene glycol

In Figure 6, the carboxylic acid O-H and amida N-H group are identified at the peak point of 3342.03 cm^{-1} . C-O ether group with a peak point of 1054.87 cm^{-1} , NO_2 at the peak 1316.21 cm^{-1} , alkene and aromatic group C = C at the peak point of 1646.91 cm^{-1} .

CONCLUSIONS

The results of this study concluded that:

- Fermentation of cassava skin using the nata method using the bacterium *Acetobacter xylinum* can be used for making bioplastics
- Bioplastics with the best characteristics, seen from the mechanical test the tensile strength, was found with the addition of 0.5% plasticizer, polyethylene glycol.
- The fermentation of cassava peel using nata method using *Acetobacter xylinum* and the addition of plasticizer 0.5% Polyethylene glycol has a characteristic as follow: tensile strength of 312.93 MPa (31.91 kgf / mm²) and elongation-to-break 5.09%.
- The results of spectrum FTIR analysis shows that bioplastic has OH, N-H, C-O, NO_2 and C=C group.

- Judging from the results of the FTIR analysis it can be said that the produced bioplastic has groups such as starch and cellulose, amide bonds on peptides and ester bonds in poly. Which are very easily degraded. This was evidenced by the results of the biodegradation test, which was 0.02 g / day.

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