© 2018 IJSRSET | Volume 4 | Issue 9 | Print ISSN: 2395-1990 | Online ISSN : 2394-4099
Themed Section : Engineering and Technology

Manufacture Dan Characterization of Bioplastic Based on Stone Banana Leaves Powder and Carboxymethyl Cellulosa (CMC) Reinforced Gum Arabic

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ABSTRACT

The bioplastic have been done by using mixture of material : stone banana leaves powder, carboxymethyl cellulosa, and gum arabic through a melt intercalation with variation of composition stone banana leaves powder : carboxymethyl cellulosa : gum arabic 80%:15%:5%, 75%:15%:10%, 70%:15%:15%, 65%:15%:20%, 60%:15%:25%, and 55%:15%:30%wt. The sample fabrication was done in three step. The first step of banana stone leaves is treated using a steam explosive method, then stone banana leaves, carboxymethyl cellulosa, and gum arabic powder sifted with particle of size 100 mesh. Then, powder is mixed until homogenous about 30 minutes with wet mixing method. Second step, the powder was homogenized then it moulded with a glass mould and cooled. Third step, bioplastic sheets has been cold, then compacted to flatten the surface with a mesh of 200 mesh at a pressure of 1 atm held for 20 minutes at 60 °C. The bioplastic sampel were characterized such as : physical properties (density, water absorption, biodegredability and functional groups) and mechanical properties (tensile strength, elongation and modulus of Young). The characterization result show that the optimum composition of stone banana leaves powder : carboxymethyl cellulosa : gum arabic 55%:15%:30%wt had the highest density 1,601 x 103 kg/m3, water absorption 10,247%, and biodegredability 99,2% for 45 days. Mechanical properties with tensile strength 35,71 MPa, elongation break 273,14% and modulus of elasticity 130,378 MPa. Thermal properties with melting point 302.45 oC whose characterization results have fulfilled the standard of SNI 7188.7: 2016. The result of its characterization has fulfilled the standard of SNI 7188.7: 2016. The results of bioplastic based stone banana leaves and CMC composite reinforced gum arabic can can be applied as packaging, and straws.

Keywords : Bioplastic, Carboxymethyl Cellulosa, Gum Arabic, Stone Banana Leaves,

I. INTRODUCTION

Plastic is a synthetic polymer material made through a polymerization process that can not be separated from the daily life of society. Based on research of each family using 1,460 plastics per year and less than 1% of plastics can be destroyed. Data from the Office of the Ministry of Environment show that each individual produces an average of 0.8 kilograms of waste per day as much as 15 percent is plastic waste. This indicates that the frequency of use of plastics as packaging material is increasing every day. This is because the plastic has advantages such as not easily broken, much lighter than other packaging materials and easily formed (sheets, pockets, or as desired design). However, conventional plastics have the disadvantages one can not decompose naturally rapidly by microorganisms present in the environment due to the constituents of the plastic itself made from petroleum products (hydrocarbons)[7]. Therefore, an attempt to reduce the consumption of plastic by synthesizing raw materials for making plastics or polymers that can be either derived by soil microorganisms called biodegradable plastics or known as bioplastics. Bioplastics are renewable plastics because their constituents come from plants such as starch, cellulose and lignin and animals such as casein, protein and lipids. Today's bioplastic is growing rapidly as a solution in dealing with non-degredable plastic problems such as conventional polyethylene (PE) plastics. Various studies have been conducted by developed countries such as Germany, Japan, Korea, the United States, the UK, the Precis and Switzerland to explore the potential of biopolymer raw materials from polyhydroxybutyrate, chitin from crustaceae, zein from corn or pullulan[3]. Indonesia is an agrarian country with potential natural resources that can be developed into cellulosic based biopolymer raw materials to produce bioplastics. Some of these herbs contain celluloses that are effective for use as plastic biopolymers such as corncobs, banana peels, sweet potatoes and so on. However, in executing to obtain the waste takes a relatively long time because waiting for raw materials or food is the first consumed[2].

So in this research, made a bioplastic from raw material of stone banana leaves and carboxymethyl cellulose (CMC) which strengthened gum arab by using melt intercalation method. The feasibility of stone banana leaves extract and CMC as the basic material of bioplastic production because stone banana leaves have cellulose content of 10.85%, hemicellulose 19.95%, lignin 18.21% and crude protein 9.24% so it can interact with matrix so it has mechanical properties the good one. Then it has the content of cuticle compounds that have a role as an anti-water and anti-evaporation agent. In addition, banana leaves also contain polyphenol compound epigallocatechin gallate (EGCG) which is an antioxidant substance and also has a polymerized tannin flovanoid substance that serves as an anti-bacterial substances[6]. CMC have hydrophilic properties composed of carboxyl groups (-COOH) and hydroxyl (-OH). Then gum arabic as a matrix due to the water absorption 0.11%, tensile strength of 18.8 N/mm^2 and modulus of elasticity of 140 N/mm^2 so as to increase the strength of bioplastik[1].

The mixture of stone banana leaves and CMC powder reinforced by gum arabic is expected to produce a bioplastic having good physical, mechanical and thermal properties in accordance with bioplastic standards of Indonesian National Standard (SNI) 7188.7: 2016. Characterization tests for bioplastics from granulated stone banana leaves and CMC-reinforced powder include: physical properties (density, water absorption and biodegredable properties), mechanical properties (tensile strength, modulus of elasticity and elongation) and thermal properties with Differential Scanning Calorimetry (DSC). It is hoped that this bioplastic will have the advantage of bioplastics already on the market, with cheaper price, practical, environmentally friendly and can create new job opportunities.

II. METHODS AND MATERIAL

A. Equipment and Materials Research

The equipments used are 100 mesh sieves, spatula, beaker glass, hot plate and magnetic stirer, oven, caliper, mixer, digital balance, mould sample from acrylic, hot press hydraulic, wayer mesh size 200 mesh, Universal Testing Machine, and Differential Scanning Calorimetry. The material used is stone banana leaves powder, carboxymethyl cellulose (CMC), gum arabic Elnasr, aquadest and 2% concentration of vinegar acid.

B. Research Variables

Research variables on the manufacture of bioplastic materials include raw material composition and characterization.

Table 1. Percentage Of Bioplastic Based On Stone Banana Leaves Powder And Cmc Reinforced Gum Arabic

Sample Code	Stone Banana Leaves Powder (%wt)	CMC (%wt)	Gum Arabic (%wt)
A	80	15	5
B	75	15	10
C	70	15	15
D	65	15	20
E	60	15	25
F	55	15	30

As for the characterization of bioplastic materials include: physical properties (density, water absorption, and biodegredable), mechanical properties (tensile strength, modulus of elasticity and elongation break) and thermal properties (melting point).

C. Research Procedures- Preparation of stone banana leaves powder

The process of making stone banana leaves powder is done by taking the leaves of banana stone with a little dark green color in the banana plantation society of Amplas. Then the banana leaves are cleaned with water and soaked for 24 hours. Cut a square-sized stone banana leaves with a size of 3 x 3 cm and soaked with 2% NaOH solution for 1 night. After it was filtered, 2% NaOH was added and fed into the autoclave at a pressure of 168.9 kPa and a temperature of 130 °C for 4 hours. Then abruptly stopped the pressure and removed the stone banana leaves from autoclave and washed with water until the pH was neutral. After that, dried banana stones that have been autoclave into the oven at 40 °C for 4 hours and blended and sieved with a size of 100 mesh.

Manufacture of Bioplastic

Bioplastic production is done by melt intercalation method. The stone banana leaves powder and CMC as filler and gum arabic as matrix are weighed by variations of composition according to Table 1 with a total mass of 10 grams. Displaced first gum arabic with 50 mL aquadest on hotplate stirer at 80 °C. Mixed powdered stone banana leaves and CMC at separate site with gum arabic solution with wet mixing at 100 mL aquadest volume while stirring then 2% vinegar added as much as 5 cc while stirring again until the mixture thickened for 30 minutes using spatula above the hotplate at 80 °C. The three ingredients are stirred by wet mixing method until uniform (homogeneous) for 10 minutes using mixer inside beaker glass. The homogeneous mixture is then fed into a square-shaped mold of acrylic. The mixture is then dried at 70 ° C and then removed from the mold so that the sheets are shaped and inserted into a 200 mesh mesh wayer and are compressed at 1 atm pressure with a 20 minute holding time at 70 °C. Released samples that have been compacted from the mold then let stand at room temperature. Bioplastics are then characterized including physical properties (density, water absorption, and biodegredable properties), mechanical properties (tensile strength, modulus of elasticity and breaking elongation) and thermal properties (DSC).

III. RESULTS AND DISCUSSION

A. Characterization of Bioplastic Physical Properties

The physical properties of bioplastic based on stone banana leaves powder and CMC-reinforced by gum arabic tested include density, and water absorption. Density is the mass measurement of each unit of material volume. Water absorption is the ability of a material to absorb water. The density test, and water absorption were performed by using Archimedes method by immersing the test sample into beaker glass for 24 hours and measured dry mass and wet mass.

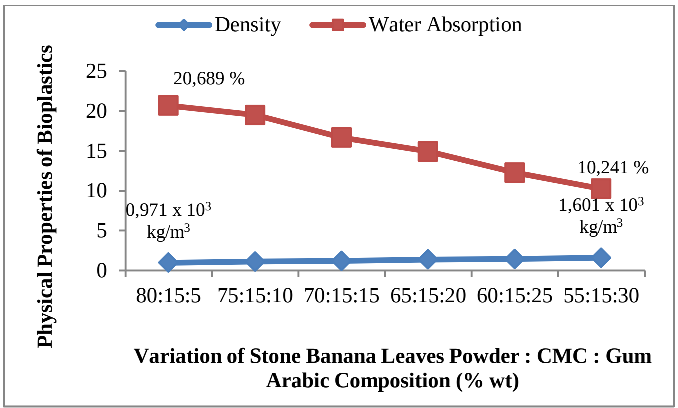

Figure 1. Test result physical properties (density and water absorption) of bioplastic based stone banana leaves powder/CMC/Gum Arabic on various variations composition

Figure 1 shows that the increased density value is proportional to the addition of gum arabic composition and the density value is inversely proportional to the water absorption. This is shown from the results of research that bioplastic optimum conditions obtained on the composition of stone banana leaves : CMC : gum arabic (55:15:30)%wt. This composition is able to produce density of 1.601 x 10^3 kg/m^3 and water absorption 10,241%. The most ugly variation conditions occurred in the composition of stone banana leaves powder : CMC : gum arabic (80: 15: 5)%wt with density of 0.971 x 10^3 kg/m^3 and water absorption 20.689%. The results of density testing and water absorption in bioplastics in this study have met the standard of conventional plastic ecogreen SNI 7188.7:2016 with density 0.95 x 10^3 kg/m^3 and water absorption of 21.5% at water-reducing temperature of 25 ^{o}C and product Enviplast brand market bioplastic with density interval (1,2 - 1,32) x 10^3 kg/m^3.

Increase in density and water absorption value not only influenced by the microstructure of the material, which includes air cavities and scratches formed during printing and cooling, but also influenced by the constituent properties of the material such as the cohesiveness cohesiveness (interface adhesions) and the roughness of the surface of the stone banana leaves powder . We can conclude that when the bioplastic conditions have a low density, it is seen that the composition of stone banana leaves powder is more dominant than that of gum arabic as a matrix so that the compounds of kutin and cellulose composed by ester group which is a combination of hydroxyl groups (-OH) and carboxyl (-COOH) having a high specific surface area is not capable of optimally bonding gum arabic bonds which cause lower polymer molecular density and water to enter easily because the hydroxyl groups are polar and hydrophilic compared to the optimum bioplastic density of the 30% gum arabic that has a higher density[8]. In addition, Gum arabic belongs to heteropolysaccharides composed of arabinous compounds consisting of aldehyde groups (C = O) and alcohol groups (-OH) and has a density of 1.485 x 10^3 kg/m^3.

B. Characterization of Biodegradable Properties

Biodegredable is a test performed to determine whether a material is either degraded or not in the environment. In this study, biodegradation tests were conducted on aerobic conditions with the help of bacteria on soil by using soil burial test method in order to know what percentage rate of bioplastic degredation with various composition variations so it can be predicted how long bioplastic will decompose by microorganisms in the soil. In this method it is only done by burying bioplastic into the controlled soil of physical and chemical properties then calculating the residual weight fraction of the bioplastics in each time unit (grams/day)[5].

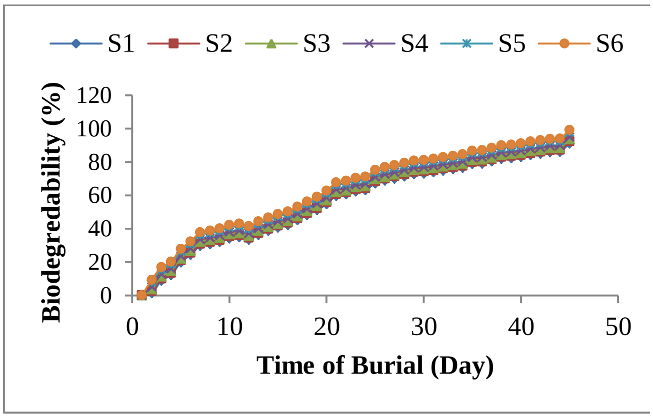

Figure 2. Test result biodegradability of bioplastic based on stone banana leaves powder/CMC/gum arabic on various variations of composition

Figure 2 shows the relationship between the length of burial time to percent biodegradable bioplastic rate referring to EN13432 standard. The results obtained for the biodegredability test buried in compost soil for 45 days showed the rate of bioplastic degredation in each composition that resulted in the biodegredability interval of 91.2% - 99.2% and fulfilled the SNI requirement that the achievement of 90% biodegredability was not more than 180 days. This is due to the bioplastic ingredients in the form of kutin, cellulose and arabinose substances which have a hydrophilic C-O ester and C = O carbonyl group-functional bonding structure which can bind water molecules from around the environment and facilitate the occurrence of degradation.

C. Characterization of Bioplastic Mechanical Properties

Tests of mechanical properties include tensile strength which is a test conducted to determine the ability of bioplastic in holding the load or mechanical force vertical given until the occurrence of broken or broken, elongatioin at break which is part of the tensile strength test conducted to determine how long the bioplastic stretch after having withdrawal force, before and after experiencing a break and modulus of elasticity is a test that aims to find out how resistant a bioplastic material undergoes strain to elastic deformation when applied vertically outward tension. Testing of mechanical properties using Universal Testing Machine (UTM) with ASTM D 882 where the results of mechanical testing include tensile strength, modulus of elasticity and the addition of breaking up as follows :

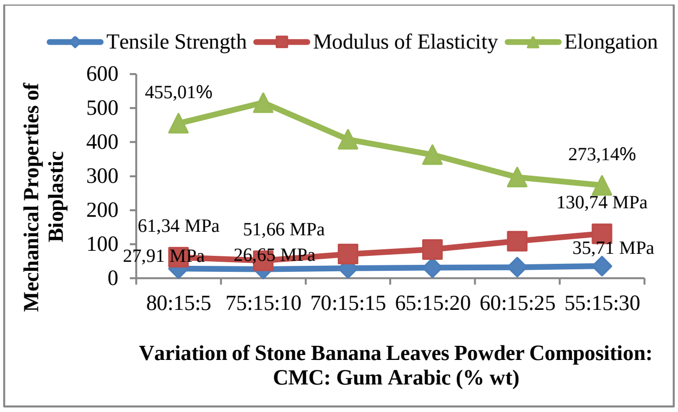

Figure 3. Test result mechanical properties (tensile strength, modulus of elasticity and elongation) of bioplastic based stone banana leaves powder/CMC/Gum Arabic on various variations composition

From the results of the observation graph above shows that the value of tensile strength and modulus of elasticity increase in proportion to the increase in the composition of the gum arabic matrix. This is shown from the result of research that optimum condition is obtained by the value of tensile strength and optimum modulus elastistas on the composition of stone banana leaves powder: CMC: gum arabic (55:15:30)%wt ie 27.91 MPa and 130.738 MPa and less optimum conditions composition of stone banana leaves powder: CMC: gum arabic (80: 15: 5)%wt yields a tensile strength of 35.71 MPa and a modulus of elasticity of 61.339 MPa. However, there is an increase and decrease in the nature of the bioplastic tensile strength starting at the composition (75:15:10)%wt with a tensile strength of 26.65 MPa and a modulus of elasticity of 51.665 MPa rising in composition (70:15:15)%wt strongly pull of 28.91 MPa and the modulus of elasticity 70,84 MPa. This is because the content of the kutin on the dried banana leaves is less than that of the banana stone in the still wet state.

The value of tensile strength and modulus of elasticity generated by the test have met the standard of conventional ecoplastic SNI 24.7 - 302 MPa and 117-137 MPa, Novamont 22 - 36 MPa bioplastics and 90 - 700 MPa, Enviplast bioplastics 12 - 18 MPa and 5.3 -6 MPa. The value of tensile strength and modulus of elasticity decreases as the filler mass increase of stone banana leaves powder resulting in the formation of hydrogen bond (H_2) and makes the spacing between bioplastic polymer chains increasingly tenuous. Where hydrogen bonds are very weak bonds, weaker than covalent bonds that lead to increased speed of viscoelastic response and molecular mobility of polymer chains in bioplastics.

While the highest percentage of elongation of breakup on the composition of stone banana leaves powder: CMC: gum arabic (75:15:10)%wt of 515.82%. However, the results of the bioplastic break-up test in this study still meet the conventional standard of ecogreen and Enviplast bioplastics with 21-220% breaking and Novamont 225 - 300% bioplastic products. The percentage rate of the break elongation is inversely proportional to the bioplastic tensile strength and the increased matrix mass of gum arabic. Decreased elongation is caused by the decrease in the number of hydrogen bonds formed by the non-optimal matrix in binding the stone banana leaves powder and CMC as filler causing the pores in the bioplastic to form, so that the viscoelasticity response decreases. Where the response causes bioplastics more rigid, hard and not elastic. The working principle of gum arabic is to form molecular interactions of polymer chains to increase viscoelastic velocity and chain mobility in polymers[4].

D. Characterization of Bioplastic Thermal Properties

The bioplastic thermal properties are determined by Differential Scanning Calorimetry (DSC) method in which the test sample will be heated from room temperature to 500 °C with a heating rate of 10 °C/min. The results of this analysis will provide information in the form of glass transition point (Tg), the point of crystallization (Tc), melting point (Tm) and decomposition point (Td) through the graph relation between heating temperature parameters (°C) on the x-axis to the energy of the given calor per minute (mW) on the y-axis is shown in Fig. 4. The test results with DSC on the optimum composition of bioplastic sample with stone banana leaves powder and CMC reinforced by gum arabic (55: 15: 30)%wt shows changes in thermal conditions through the same six stages which is shown by the peaks generated by the DSC tools ie valley peaks and hilltops. Where the peak changes by the DSC occur as a result of changes and chemical reactions followed by temperature changes in the test sample. The chemical reactions that occur in DSC devices are exothermic and endothermic reactions.

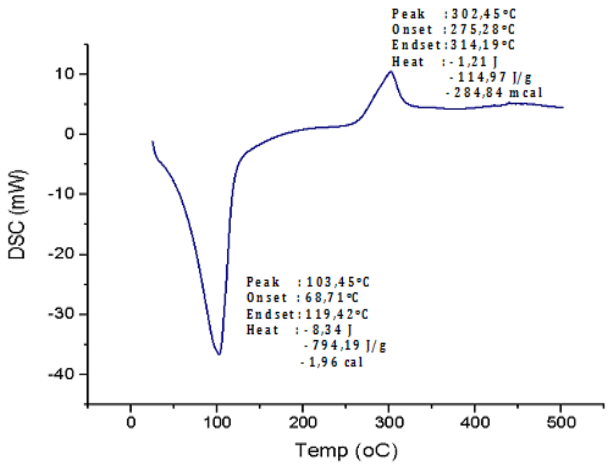

Figure 4. Thermal analysis (DSC) of bioplastic stone banana leaves powder/CMC/gum arabic on variation (55: 15: 30)%wt

Based on the graph of Figure 4 shows the results of the analysis of the bioplastic thermal properties on the variation of composition of stone banana powder, CMC, and gum arabic (55:15:30)%wt given by the first maximum peak for the process of thermal change of absorber material (bioplastic) a temperature of 68.71 °C - 119.42 °C produces a valley peak that informs the point of crystallization (Tc) in which a crystalline polymer at a temperature of 103.45 °C then occurs an endothermic process in which the composition of the absorber material begins to absorb heat by a large amount of heat energy absorbed ie

8.34 Joules. In the endothermic process the material of the absorber material undergoes a rubberized deformation at a glass transition point (Tg) of 119.42 °C and the highest peak for an exothermic reaction at a temperature of 275.28 °C - 314.19 °C with a thermal energy released equal to 1.21 Joule yields a melting point of 302.45 °C. Where the melting point of Enviplast bioplastic is 140 - 160 °C.

IV. CONCLUSION

In the experiment, bioplastic was obtained based on banana leaf powder and CMC as filler with gum arabic adhesive as matrix which was made using melt intercalation method. This bioplastic has the potential as an alternative packaging material. Bioplastics that have been made have advantages, between: strong, elastic, heat resistant, waterproof, and environmentally friendly. The optimum condition of bioplastic (composition of banana leaf powder, CMC and gum arabic 55: 15: 30 wt) was able to produce density 1.601 x 103 kg / m3, water absorption 10,241%, biodegredability 99.2% for 45 days, tensile strength 27.91 MPa, elongation 273,14 elastic modulus 130,738 MPa and melting point 302.45 oC. Thus, this bioplastic has the potential to be applied as a simple packaging technology that can be used to answer the problem of plastic waste that can not decompose by microorganisms.

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