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PREPARATION, CHARACTERIZATION, FUNCTION, AND DEGRADATION OF BIOPLASTICS FROM VEGETABLE WASTE COATING WITH POLYEUGENOL

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ABSTRACT

This research aimed to prepare, characterize, function, and degrade bioplastics from vegetable waste via an eco-friendly method. The production of bioplastics using CH_3COOH 1 M and blending with Polyvinyl Alcohol (PVA) in water, is easily saleable and does not involve the use of hazardous or organic chemicals, which raises environmental issues. Vegetable wastes like moringa (M), water spinach (WS), and green spinach (GS) blended with PVA were used to create free-standing, flexible bioplastic films. The result showed these bioplastics have tensile strength from 2.63-6.70 MPa, elongation from 5.33-23.5 %, and modulus young from 13.67-51.30 MPa. Bioplastic's antioxidant properties belong to low antioxidants and coating bioplastic with polyeugenol can be increased antioxidant properties. Bioplastic from WS vegetable waste/PVA/Polyeugenol (PEU) can be used to increase the self-life of apple fruit. This research also examines the function and degradation of bioplastic from water spinach and its degradation. All these bioplastics were potentially applicable as active packaging material.

Keywords: Bioplastics, Vegetable Waste, Polyeugenol, Function, Degradation, Active Packaging Material.
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INTRODUCTION

The use of plastic has expanded and touched almost all lines of life. We can find various products and equipment made from this material because it is considered more economical, not easily broken, flexible, and lightweight. One example of a product made from plastic that is most often used by the public is plastic bags and utensils. Indonesia's per capita plastic consumption is around 10 kg/capita/year. Compared to other ASEAN countries; the amount of plastic used in Indonesia is still lower. Malaysia, Singapore, and Thailand reach figures above 40 kg/capita/year. Although low, the potential for increasing demand is still quite large, which is around 4.6 million tons/year with a growth of around 5% per year.[1] The use of plastics as packaging materials and other equipment has caused environmental problems. Plastic waste is increasingly piling up, because plastic waste is not easily destroyed either by rainy weather and the sun or by microbes that live in the soil, thereby increasing environmental damage such as soil pollution.[2-6] This is because the plastic that has been used so far comes from petroleum and natural gas. Currently, these basic materials are starting to experience a reduction and cannot be renewed.[7] One solution to overcome this environmental problem is to develop biodegradable plastic materials (bioplastics) or plastics that are more easily decomposed naturally by microorganisms into simple compounds that are environmentally friendly. The development of bioplastic materials using vegetable waste is expected to better overcome environmental pollution. The materials used to make biodegradable plastics include polymer compounds found in plants such as starch, cellulose, and lignin, as well as in animals such as casein, chitin chitosan, and so on.[8] The description underlies this research and the existence of this research is expected to be a reference on the utilization of waste vegetable moringa (M), water spinach (WS), and green spinach (GS) mixed with PVA as the basic material for making bioplastics. On the other hand, cloves (*Syzygium aromaticum*) are the aromatic dried flower buds of an evergreen tree in the family *Mytraceae*. Cloves are used in Ayurveda called Lavang in India. Indonesia is among the top oil producers in the world. Furthermore, about half of the clove leaf oil used globally is consumed in Indonesia. The percentage of eugenol, also known as 3-(4-hydroxy-3-methoxyphenyl) propene, is set at between 80 and 90 percent by weight in oil.[9-14] Furthermore, the potential for polyeugenol (PEU) material for the development of more useful compounds. Bioplastic vegetable waste from Moringa (M), Water Spinach (WS), and Green Spinach (GS) can be used as raw material to produce bioplastic to replace the role of conventional plastics. In this research, its function was also tested to increase the shelf-life of apple fruit and their degradation in the soil.

EXPERIMENTAL

Material and Methods

The materials to be used in this research are M waste, WS waste, GS waste, CH_3COOH glacial, polyvinyl alcohol, aquadest, 2,2-diphenyl-1-pikrilhydrazil (DPPH), ethanol, ethyl acetate, apple fruit, soil. Besides, PEU was prepared by reacting eugenol with H_2SO4-CH_3COOH. [11]Bioplastics were prepared by hydrolysis vegetable waste with 1 M CH_3COOH and blending with PVA and coating using PEU.[12]

General Procedure

Antioxidant Test

A blank solution was made by pipetting 1 mL of 0.4 mM DPPH solution, then the volume was made up to 5 mL with methanol p.a. After that, it was left for 30 minutes in a dark place, and the absorption of the solution was measured with a UV-Vis spectrophotometer at a wavelength of 400-600 nm. The mother liquor of the bioplastic extract was prepared by weighing 0.01 g and then dissolving it with 10 mL of ethanol. The mother liquor with known concentration was made in 5 concentration series, namely 20, 40, 60, 80, and 100 ppm. Take 1 mL of each solution and then add 3 mL of DPPH solution, the mixture is shaken and left for 30 minutes at room temperature and in a room protected from sunlight. The absorbance (A) was measured at the maximum wavelength with a UV-Vis spectrophotometer. Furthermore, the percentage of inhibition (inhibition) is calculated with the equation:

$$\text{Inhibition } (\%) = \frac{\text{Abs control} - \text{Abs Sample}}{\text{Abs control}} \times 100$$

Where:
Abs control = DPPH + methanol Absorbance
Abs sample = DPPH absorbance + sample
Furthermore, the value of % inhibition was used to calculate the IC_{50} value (ppm).

Mechanical Test

The bioplastic film that has been made is cut to a predetermined shape according to the Universal Testing Machine then the value of the area of the bioplastic film and the tensile strength value, elongation and modulus young measured with the Universal Testing Machine

Coating Bioplastic With PEU

While the PEU was dissolving in ethyl acetate, bioplastic was applied to the preparation. On one side of the bioplastic, both coats were sprayed using the spray technique using a hand sprayer. Regarding the weight of the bioplastic, different concentrations of PEU (10 wt%, 20 wt%, and 30 wt%) were added to the substrate. A reference sample that was treated with only pure water was also created in addition to the tissues treated with active compounds. Each sample had the same quantity (20 mL) of ethyl acetate. The coating was applied 10 cm away from the bioplastic's surface to do so uniformly.

Application of Bioplastics in Apples Fruit

Prepare 5 beakers, then put the cut apple into each beaker. Next, the beakers containing apples are coded A, B, C, D, and E, where beaker A is without covered with plastic, glass B is covered with plastic, and glass C is covered with bioplastic/PVA/PEU. Then see the changes for 16 days and will be monitored every 2 days.

Bioplastic Degradation

Bioplastic is cut into 2×2 cm2 sizes and weighed. Then immerse it in the ground to a depth of 5 cm. The soil was watered once every day for 16 days. The bioplastic is then taken from the soil every day, washed, and dried in an oven at 50°C until dry. Then weighed, the degradation percentage is calculated following the equation:

$$\text{Degradation } (\%) = \text{Mi} - \frac{\text{Ma}}{\text{Mi}} \text{x } 100\%$$

Where, Mi is the initial mass of the plastic before the degradation test is carried out, and Ma is the final mass of the plastic after the biodegradation test is carried out.

Detection Method

UV-Vis spectrophotometer (Shimadzu FTIR-8100) measured IC_{50}. Tensile strength, elongation, and modulus young were used universal testing machine (Instron).

RESULTS AND DISCUSSION

Mechanical Properties

Mechanical properties are influenced by the amount of content of the components that make up bioplastic. The purpose of testing the mechanical properties is to determine the characteristics of the best mechanical properties of bioplastics with variations in synthesis temperature of 30^0C, 50^0C, and 70^0C.

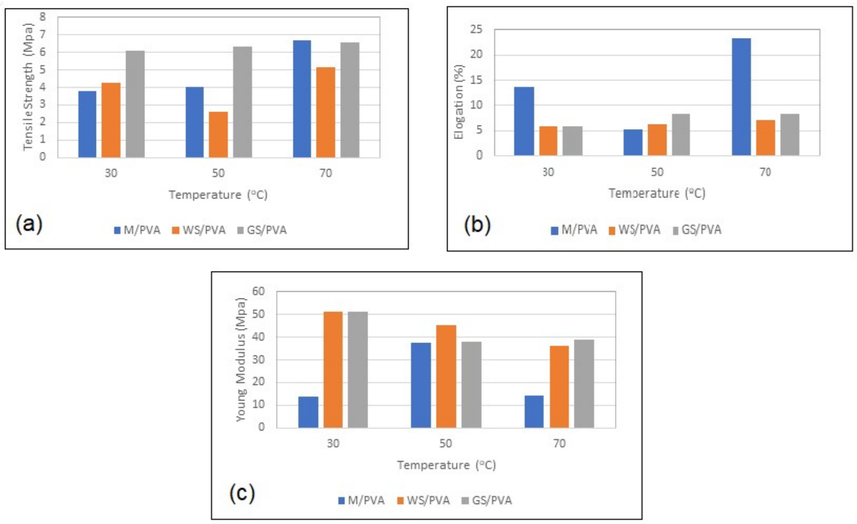


Fig.-1: Mechanical Properties of M/PVA, WS/PVA, and GS/PVA

The molecular size and number of hydroxyl functional groups of plasticizers as well as compatibility with polymers can affect the interaction between plasticizers and polymers. In Fig.-1 (a) the tensile strength of bioplastics increases with increasing synthesis temperature, it can be concluded that the optimum temperature for the tensile strength of bioplastics from moringa vegetable waste is at a temperature of 70^0C. The tensile strength values of bioplastics with variations in temperature at 70^0C in this study were 6.70 Mpa, 5.16 Mpa, and 6.58 Mpa, which means that bioplastics from vegetable waste in this study which is close to the value of the tensile strength of bioplastics in the research conducted which stated that the tensile strength of bioplastics from a mixture of pure cellulose and PVA ranged from 8.8 to 16.4 Mpa and with the tensile strength values of glucomannan and starch bioplastics, namely 2.12 Mpa – 6.17 Mpa.[14] Figure-1 (b) shows the highest elongation value of 23.50% of bioplastics at a synthesis temperature of 70 ^{0}C.

It can be concluded that the optimum temperature for the value of bioplastic elongation from moringa vegetable waste is at a temperature of 70^0C which follows a study conducted that stated that the elongation value of bioplastics from orange peel and spinach stems ranged from 9 -23%.[15] Young's modulus is obtained from the ratio of the tensile strength to the percent elongation. Figure 5 shows the highest Young's modulus value of 51.30 MPa from GS vegetable waste bioplastic at a temperature of 30^0C. Tensile strength and elasticity are directly correlated, but elongation and elasticity are inversely correlated[4], but in this study the data obtained were inconsistent.

This is thought to occur because the plasticizer used does not dissolve completely in water so the compatibility is less than optimal which affects the resulting Young's modulus value. The elasticity value of bioplastics from vegetable waste is 16.24 MPa-51.30 MPa, which follows the research that stated that the value of the bioplastic Young's modulus from orange peel was 25.33 Mpa.[16]

Coating of Bioplastic from WS with PEU

Mechanical Properties

From Table-1 it can be seen that the bioplastic coating of waste with polyeugenol can increase the tensile strength and Young's modulus but decrease the elongation value. The addition of PVA significantly increased tensile strength, elongation, and Young's modulus where the values were similar to other bioplastics.

Table-1: Effect of PEU on Mechanical Properties of Bioplastics from WS Vegetable Waste

Mechanical Properties	WS (Mpa)	WS/PEU 10% (Mpa)	WS/PEU 20% (Mpa)	WS/PEU 30% (Mpa)	WS/PVA/PEU 30 %- (Mpa)
Tensile Strenght	0.16	0.30	0.23	0.24	2.37
Elogation	4.67	3.83	3.50	3.00	11.00
Modulus Young	1.73	4.26	3.32	3.98	10.80

Antioxidant Activity

The DPPH technique was used to conduct the antioxidant activity test. The IC50 value, or the concentration of the test substance needed to decrease DPPH radicals by 50%, is the metric used to assess antioxidant activity using the DPPH technique.

Table-2: Antioxidant Activity of Bioplastics Using the DPPH Method

Vegetable Waste	IC_{50} Temperature 30 ºC (µg/mL)	IC_{50} Temperature 50 ºC (µg/mL)	IC_{50} Temperature 70 ºC (µg/mL)
Moringa	161.06	232.13	159.18
Water Spinach	261.32	389.29	288.57
Green Spinach	2,790.00	2,454.89	248.09

The IC_{50} value data were obtained successively for bioplastics from M vegetable waste at a temperature of 30ºC; 50 ºC; 70 ºC is 161.06 g/mL; 232.13 g/mL; 159.18 g/mL. It shows that all samples have weak antioxidant values. For bioplastic from WS waste, at temperatures 30; 50 ºC; and 70 ºC 261.06 g/mL; 389.29 g/mL; and 288.57 g/mL showed that all samples had very weak antioxidant values. In this study, the addition of polyeugenol into the resulting bioplastic from WS was done by coating using a hand sprayer, this method includes a simple way to coat bioplastics with polymers.

Table-3: The Effect of Polyeugenol on Bioplastic Antioxidants from Water Spinach Vegetable Waste.

Sample	IC_{50}(µg/mL)
Control	97.35
Polyeugenol 10%	47.14
Polyeugenol 20 %	6.51
Polyeugenol 30%	95.06
Polyeugenol 30%-PVA	33.2

This is because the PEU structure contains an OH group so it is easier to bind with water. The addition of active ingredients that have antioxidant and antibacterial properties can increase their antibacterial and antioxidant properties. PEU has strong antioxidant and antibacterial activity.[11] In this study, PEU was used to increase its antioxidant properties and at the same time as an antibacterial and not reduce its tensile strength. From Table-4 it can be seen that the addition of PEU significantly increased its antioxidant properties from strong antioxidant to very strong. In the presence of PEU, of course, the resulting bioplastic has antibacterial properties.

Bioplastic was applied to apples to determine the ability of bioplastic to protect the product. In this study, the color of the apples that were stored covered changed from day 2 to day 6 (Fig.-3). When stored, uncoated apples and apples coated with petroleum-based plastic showed a brownish color change compared to apples coated with PEU 10%, 20%, and 30%. Colourless fruit may experience a browning reaction. In research that has been carried out, bioplastic coated with PEU can protect apples because it has antioxidant and antibacterial activity. According to Rahim[11], PEU compounds have quite strong antioxidant activity.

Function of Bioplastic WS Vegetable Waste/PVA/PEU

Polyeugenol contains phenolic compounds, the presence of phenolic compounds in PEU will cause these compounds to have good antioxidant activity. This happens because phenolic compounds are known to have hydroxyl groups (OH). Practically, the hydrogen atom from the phenolic group will be donated to the radical compound to be stabilized. In addition, this research has produced PEU with a high molecular weight, so its antioxidant properties are higher than eugenol.

Bioplastic Degradation

Lastly, a study was carried out to ascertain the biodegradation process of the selected materials (WS/PVA-10% PEU, WS/PVA-20% PEU, and WS/PVA-30% PEU) and their control (WS/PVA-0% PEU) in soil. Figure-3 displays the findings. Every sample in the experiment had its weight loss monitored, and at the beginning of the investigation, there were significant differences between the coated and uncoated materials (Fig.-2), even though the coated materials had greater material integrity and, by the end of the investigation, were clearly showing slower biodegradation. All of them degraded in the soil rapidly overall; 80% of the coated materials biodegraded in seven days, and 100% of the controls in just five days.

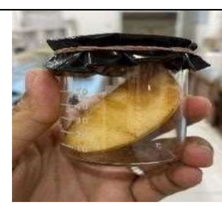
	Application of Bioplastic to Increase Self-Life of Apple Fruit		
	1th Day	4th Day	6th Day
Without plastic			
Commercial plastic			
Bioplastic WS Vegetable Waste/PVA			
Bioplastic WS Vegetable Waste/PVA/PEU 10%			
Bioplastic WS Vegetable Waste/PVA/PEU 20%			
Bioplastic WS Vegetable Waste/PVA/PEU 30%			

Fig.-2: Application of Bioplastic WS Vegetable Waste/PVA/PEU

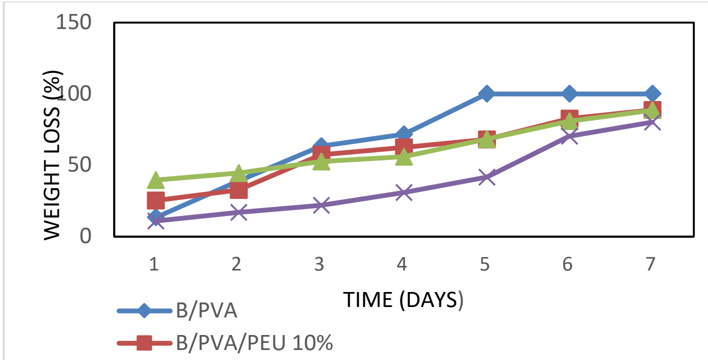


Fig.-3: Degradation of Vegetable Waste Bioplastics in Soil

CONCLUSION

Based on the results and discussion, vegetable wastes like moringa, water spinach, and green spinach blended with PVA were used to create free-standing, flexible bioplastic films. The result showed these bioplastics have tensile strength from 2.63-6.70 MPa, elongation from 5.33-23.5 %, and modulus young from 13.67-51.30 MPa. However, Bioplastic's antioxidant properties belong to low antioxidants. Blending with PVA and coating with PEU can increase antioxidant properties and become a very strong antioxidant. WS/PVA/PEU can be used to increase the shelf life of apples and can be degraded easily in the soil. All these bioplastics were potentially applicable as active packaging material.

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CONFLICT OF INTERESTS

The authors declare that there is no conflict of interest.

AUTHOR CONTRIBUTIONS

All the authors contributed significantly to this manuscript, participated in reviewing/editing, and approved the final draft for publication. The research profile of the authors can be verified from their ORCID ids, given below:

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