Journal of Vocational Studies on Applied Research

The Effect of Differences in the Use of Flour and Plasticizers in Making Biodegradable Plastic on the Physical Characteristics of Beeswax

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Abstract *– Plastic is one of the objects that is necessary in daily activities. This is because plastics make it easier for humans to package goods. Its elastic and easy-to-use properties make plastic widely used by humans. However, on the other hand, plastic has properties that are difficult to decompose (non-degradable), so it is one of the biggest generators of waste that pollutes the environment. Biodegradable plastic or commonly called bioplastic, can be an alternative product to reduce plastic waste because it is easily decomposed and more environmentally friendly. This research examines the influence of the type of starch (avocado seed flour, mango seed flour, banana peel flour, cassava peel flour) and plasticizers (glycerol, sorbitol, and polyvinyl alcohol) with beeswax additives on the characteristics of the resulting bioplastic. The best tensile strength value was found in a sample of mango seed flour, PVA-type plasticizer, and 0.04 grams of beeswax, namely 11.35 MPa. The best elongation value was found in samples of mango seed flour, glycerol-type plasticizer, and 0.04 grams of beeswax, namely 67%. The best percentage degradation value is bioplastic using cassava peel starch. The best swelling index value was found in samples of avocado seed flour, sorbitol-type plasticizer, and 0.1 grams of beeswax, namely 20.16%. In terms of water content values, all samples are in accordance with SNI, namely below 20%.*

Keywords: *plastic; biodegradable; beeswax; starch; plasticizer*

Received: August 22, 2024 Revised: September 14, 2024 Accepted: December 12, 2024

Doi: http://dx.doi.org/10.14710/jvsar.v6i2.24257
[How to cite this article: Yustianto, A.H., Sully, D.A., Fauzi, A.D., and Amalia, R. (2024). The Effect of Differences in the Use of Flour and Plasticizers in Making Biodegradable Plastic on the Physical Characteristics of Beeswax. Journal of Vocational Studies on Applied Research, 6(2), 14-21. doi: http://dx.doi.org/10.14710/jvsar. v6i2.24257]

INTRODUCTION

Waste is a by-product of human activities that is no longer used, originating from the domestic and commercial sectors, ranging from households to industry. Plastic waste is a type of waste that is difficult to decompose by soil microorganisms (non-biodegradable), so it has a negative impact on the environment. According to the Environmental Statistics Agency (2019), in 2019 the accumulation of plastic waste increased to 67 million tonnes. Data from the Ministry of the Environment shows that each individual produces an average of 0.8 kilograms of waste per day, and 15 percent of this is plastic waste. This problem makes humans innovate in making plastic packaging that has biodegradable properties in order to minimize the environmental impact that occurs.

Bioplastics are plastics that come from renewable materials and can decompose back into nature. Bioplastics are made from biomass with or without modification, such as starch, protein, and cellulose (Nguyen et al., 2022). Starch has the potential to be used as raw material for biodegradable plastic because it is cheap, easy to obtain, and renewable (Santana et al., 2018). Starch itself is a polymer in the form of long chains of glucose found in plant tissue (Nisah, 2018). Starch consists of amylose and amylopectin, where the amylose molecules are smaller and have an unbranched structure, while the amylopectin molecules are larger with a branched structure and form a double helix. (Lintang et al., 2016). Amylopectin plays a role in the stability of biodegradable plastics, while amylose plays a role in hardness or compactness (Niken & Adepristian, 2013). Starch can be obtained from various plants or agricultural materials such as corn, rice, cassava, potatoes, nuts, seeds, and many more starches produced by plants (Indarti et al., 2022). In this research, flour from agricultural waste containing starch was used, namely avocado seed flour, mango seed flour, banana peel flour, and cassava peel flour. Study Ginting et al. (2015) got a starch content in avocado seeds of 24.20%, mango seeds contain 58% starch (Kusmiyati et al., 2019). The starch content in banana peels is 12.78% (Emaga et al., 2007). Cassava skin contains 59% starch or starch (Mudaffar, 2021).

Making bioplastics from starch alone still has shortcomings, such as being easy to tear, so it is necessary to add plasticizers to improve its properties (Robiah, 2020). Plasticizers are substances that can increase the elasticity and flexibility of biodegradable plastics. Glycerol and sorbitol are the softening agents most often used in making bioplastics (Vieira et al., 2011). In this research, glycerol, sorbitol, and polyvinyl alcohol were used as plasticizers with the aim of determining the effect of the differences between the three plasticizers used on the properties of the bioplastics produced. On research by Purba et al. (2019) related to the manufacture of cornstarch bioplastic using glycerol plasticizer, the best results were tensile strength of 2.2 MPa, elongation of 22%, swelling of 57%, and degradation rate of 8 days. Meanwhile, with the sorbitol plasticizer, the best results were obtained for tensile strength of 1.2 MPa, elongation of 28%, swelling of 97%, and degradation rate of 8 days. Then on to research Coniwanti et al. (2014), in making bioplastics from corn starch with the addition of chitosan and glycerol, the best performance was obtained, namely tensile strength values of 3.92 MPa, elongation of 37.92%, and swelling of 26.78%. Meanwhile, in research, Purnavita & Anggraeni (2019) in making bioplastics made from glucomannan-PVA polyblend with the addition of beeswax and glycerol, it had the best tensile strength results of 0.46 kg/mm2 and elongation percentage of 484%. The addition of plasticizers, apart from increasing flexibility, also increases permeability so that the packaging is deemed unable to protect the product optimally from the effects of CO_2, O_2, and water vapor. Purnavita & Anggraeni (2019), so it is necessary to add other substances that can reduce permeability.

Beeswax is a substance that can reduce the permeability of starch-based bioplastics so that it can increase the function of plastic as packaging to protect the product inside. The addition of beeswax also increases the tensile strength of the plastic (Mujahid, 2019). Beeswax consists of lipid components sourced from honey bee hives, so it is abundantly available, has an affordable price, including food grade, and is easily degraded (Purnavita & Anggraeni, 2019). Beeswax is a white solid that has a melting point at a temperature of 61- 65ºC.

Based on the background above, this research aims to determine the effect of different types of starch and plasticizer on the characteristics of biodegradable plastic made from additional beeswax. In this research, several types of starchy flour were used with the addition of plasticizers. The experiment was carried out using a completely randomized design (CRD) with three factorials. The first factor is the type of starchy flour (avocado seed flour, mango seed flour, banana peel flour, and cassava peel flour). The second factor is the type of plasticizer (glycerol, sorbitol, and polyvinyl alcohol). The third factor is the beeswax concentration. The parameters measured include swelling value, degradability value, tensile strength, elongation, and water content.

MATERIALS AND METHODS

Materials

The materials used were acetic acid, EM4 solution, distilled water, avocado seed flour, banana peel flour, mango seed flour, avocado seed flour, cassava peel flour, glycerol stabilizer, sorbitol, and polyvinyl alcohol.

Procedure for Making Bioplastic Films Made from Natural Starch

A total of 5 grams of various natural starchy flours (avocado seed flour, banana peel flour, mango seed flour, cassava peel flour) is dissolved in 50 ml of distilled water and filtered. Then the filtrate is added with acetic acid 5%, plasticizer as much as 8% of the starch weight, and beeswax (0.04 gr and 0.1 gr). Then the mixture was stirred for 35 minutes at 70ºC using a magnetic stirrer until gelatin formed. Carried out on every variation of flour and plasticizer variation. Next, the mixture is poured onto the silicone and dried in an oven at 55ºC for 5-7 hours until the bioplastic can be removed from the mold.

Tensile Strength Test

The tensile strength test, or what is usually called tensile strength is a test related to strength that is used to determine the durability of a material. It is a measure of the amount of force that a material can withstand before it breaks or cracks. Tensile strength testing is carried out by measuring the force required to pull a material until it breaks or cracks. This test is carried out using a tensile testing machine or tensometer, which can measure the force applied and the length of the material being tested

Tensile strength testing is a test to determine the physical characteristics and durability of the bioplastics that are formed (Purnavita & Anggraeni, 2019). The tensile strength value formed can be calculated using the following equation:

$$Tensile\ Strength = \frac{Maximum\ Force}{Cross\ Sectional\ Area} \quad (1)$$

Elongation Test

The elongation test, also known as the length increase test, is a test that aims to determine the value of the maximum length increase produced by a bioplastic after additional force is applied (Purnavita & Anggraeni, 2019). The elongation value is calculated using the following equation:

$$Elongation = \frac{Increased\ Lenghth}{Initial\ Length}\ x\ 100\% \quad (2)$$

Biodegradation Test Analysis

The environmentally friendly plastic that has been made is tested for its biodegradability with the help of Effective Microorganisms or EM4 bacteria, which are composting bacteria. Biodegradation ability is seen based on the length of degradation time by EM4 microorganisms. The biodegradation test was carried out by placing the film sample on a piece of soil, which was then added with 10 ml of EM4 and left to degrade (Mujahid, 2019). Percent weight loss can be calculated using the following formula.

$$\%W = \frac{(Initial\ Weight - Final\ Weight)}{Final\ Weight}\ x\ 100\% \quad (3)$$

Swelling Test

The swelling test is a test that aims to determine the ability of plastic to swell when placed in a solution. In this study, the swelling test was carried out by weighing the bioplastic before soaking, and then the sample was soaked in water for approximately 30 minutes (Mujahid, 2019).

Water Content Measurement

The sample was cut into small pieces and then weighed 1 gram in a porcelain cup that had previously been weighed to determine the weight. The cup is placed in the oven for 5 hours at a temperature of 100°C – 105°C then cooled in a desiccator until it reaches a constant weight (Purnavita & Anggraeni, 2019). Weight loss will be calculated as a percentage of water content and calculated using the formula:

$$WC\ (\%) = \frac{Initial\ Mass - Constant\ Mass}{Initial\ Mass}\ x\ 100 \quad (4)$$

RESULTS AND DISCUSSION

Tensile Strength and Elongation of Bioplastics According to Treatment Combinations

Tensile strength is the maximum value that can be obtained when a tensile test is carried out. Bioplastics that have high tensile strength values are able to provide protection to the products they package (Hasanah et al., 2017). Table 1 shows the results of the tensile strength analysis and the results of the bioplastic elongation analysis. The best results for tensile strength values were obtained for bioplastics with variables such as manga seed flour and PVA plasticizer type, and 0.04 grams beeswax with a tensile strength value of 11.35 Mpa. The best elongation percentage value was obtained in bioplastic with the addition of manga seed flour, with the addition of 0.04 beeswax, and the addition of glycerol plasticizer with an elongation percentage value of 67%.

The results obtained in Table 1 show that the best tensile strength analysis results were obtained when using mango seed flour, which obtained an average tensile strength value of 4.33 Mpa, which is in accordance with previous research conducted by Robby et al. (2021), where the value of starch content greatly influences the tensile strength of the bioplastic that is formed. The higher the starch content used, the lower the tensile strength value of the bioplastic. Meanwhile, the best elongation value was obtained from cassava peel flour, where the average elongation percentage value was 31.45%, according to Zulisma Anita et al., (2013), the presence of starch in bioplastics influences the elongation value significantly. In general, the higher the starch content, the higher the elongation value of bioplastics. This is because starch has a flexible molecular structure and is easily decomposed.

Table 1. Tensile Strength and Elongation Result

	Plasticizers	Bees wax (gr)	Tensile Strength (MPa)	Elongation at Break (%)
Mango Seeds	Glycerol	0.04	1.01	67.90
		0.10	0.61	27.90
	Sorbitol	0.04	2.14	43.00
		0.10	1.18	41.70
	PVA	0.04	11.35	3.00
		0.10	9.04	2.70
Banana peel	Glycerol	0.04	0.70	36.00
		0.10	0.30	43.00
		0.04	1.74	31.30

	Sorbitol	0.10	2.18	43.30
	PVA	0.04	8.07	2.90
		0.10	6.55	2.50
Cassava Skin	Glycerol	0.04	0.32	56.00
		0.10	0.46	55.00
	Sorbitol	0.04	1.38	25.70
		0.10	1.37	31.80
	PVA	0.04	3.67	9.30
		0.10	3.14	10.90
Avocado Seeds	Glycerol	0.04	0.52	45.00
		0.10	0.33	43.00
	Sorbitol	0.04	2.24	32.20
		0.10	1.70	36.30
	PVA	0.04	2.26	9.70
		0.10	1.19	8.60

Choosing the right type of plasticizer can increase the tensile strength value of bioplastic. Based on the data, the largest tensile strength value was obtained when using PVA plasticizer, where the average value was 3.918 Mpa. Meanwhile, the best elongation value was obtained when using glycerol plasticizer where the average elongation value was 46.84%. Similar results were reported by Isotton et al. (2015) where the use of a glycerol type plasticizer produces a very small tensile strength value but with elongation which is higher than films with PVA or sorbitol type plasticizers, while when using PVA type plasticizers the value is obtained tensile strength which is very large compared to usage PVA or glycerol plasticizer but with very low elongation values. According to Sagar et al. (2012) in general, materials with high tensile strength will have low elongation & vice versa.

Table 1 shows that there are two variables for using beeswax with different concentrations, where the best tensile strength and elongation values were obtained using the variable 0.04 grams of beeswax with an average tensile strength value of 3 Mpa and elongation of 43%. According to Purnavita & Anggraeni (2019), in general the addition of beeswax at low concentrations can increase the tensile strength and elongation values of bioplastics, this is because beeswax acts as a reinforcement, filling gaps and cavities in the structure of bioplastics, but the use of beeswax with excessive concentrations can reduce the level of beeswax. stiffness, durability, and flexibility value of the bioplastic formed.

Percent Degradation of Bioplastics According to Treatment Combinations

Degradation analysis is aimed at analyzing the ability of microorganisms to degrade bioplastics. Bioplastics resulting from research were tested for degradation time analysis, testing using EM4 bacteria as microorganisms. EM4 bacteria are bacteria that are often used in the fermentation process of soil organic matter. EM4 bacteria will produce enzymes to degrade bioplastics by breaking polymer chains into monomers (Yuniwati et al., 2017). EM4 contains fermentation bacteria from the genus Lactobacillus, fermentation fungi, photosynthetic bacteria Actionomycetes, phosphate solubilizing bacteria and yeast Saputro & Ovita (2017).

Table 2 shows the effect of using a combination of different methods on the degradation ability of bioplastics. Based on the results obtained, it shows the best % degradation value when using cassava peel flour. This is due to the use of starch content in cassava skin flour which is not too high, namely 30%. This is supported by Saputra & Supriyo (2020) where in general the use of excessive starch content will cause an increase in the number of glycosidic bonds found in bioplastics, where these glycosidic bonds have high resistance to degradation processes caused by microorganisms.

Table 2. Percent Degradation of Bioplastics Result

	Plasticizers	Beeswax (grams)	Degradation (%)
Mango Seeds	Glycerol	0.04	100.00
		0.10	83.09
	Sorbitol	0.04	65.01
		0.10	45.93
	PVA	0.04	87.90
		0.10	100.00
Banana peel	Glycerol	0.04	100.00
		0.10	76.01
	Sorbitol	0.04	61.03
		0.10	51.04
	PVA	0.04	91.03
		0.10	65.01
Cassava Skin	Glycerol	0.04	100.00
		0.10	100.00
	Sorbitol	0.04	100.00
		0.10	85.90
	PVA	0.04	100.00
		0.10	100.00
Avocado Seeds	Glycerol	0.04	100.00
		0.10	100.00

Sorbitol	0.04	73.76
	0.10	47.09
PVA	0.04	100.00
	0.10	100.00

Different types of plasticizer use provide different results. Where the highest average % degradation value was obtained when using the plasticizer Glycerol, namely 94.88% compared to PVA of 92.99%, and the lowest degradation value was obtained when using the plasticizer sorbitol, 66.22%. These results are supported by previous research where in this research it was found that bioplastics using glycerol plasticizers were more easily degraded because glycerol more easily absorbs water content compared to other types of plasticizers, which makes it easier for microorganisms to degrade bioplastics (Lusiana et al 2019).

The addition of a greater concentration of beeswax causes an increase in the duration of the bioplastic degradation rate. This is because the addition of beeswax causes a reduction in the ability of bioplastic to absorb water content, which can complicate the degradation process of bioplastic itself Khanzadi et al., (2015).

Swelling Index According to Treatment Combination

The swelling test values are shown in Table 3 where it is known that the best swelling value was obtained in the sample combination of avocado seed flour with the addition of the plasticizer sorbitol and the addition of 0.1 gr beeswax where the swelling percentage value was obtained at 20.16% which has the best water resistance compared to others.

Table 3. SwelIing Index and Water Content Result

	Plasticizers	Beeswax (grams)	Swelling (%)	Water content (%)
Mango Seeds	Glycerol	0.04	39.48	14.44
		0.10	37.11	14,19
	Sorbitol	0.04	36.33	13.00
		0.10	25.62	13.85
	PVA	0.04	29.34	11.70
		0.10	26.38	10.03
Banana peel	Glycerol	0.04	32.71	17.61
		0.10	31.41	17.75
	Sorbitol	0.04	31.03	13.82
		0.10	25.98	13.03
	PVA	0.04	35.98	6.42
		0.10	27.07	5.20
Cassava Skin	Glycerol	0.04	34.51	16.79
		0.10	30.45	16.27
	Sorbitol	0.04	48.83	14.20
		0.10	26.87	14.62
	PVA	0.04	34.78	10.39
		0.10	32.55	10.29
Avocado Seeds	Glycerol	0.04	42.80	14.79
		0.10	35.69	14.27
	Sorbitol	0.04	26.84	13.73
		0.10	20.16	14.56
	PVA	0.04	41.13	10,11
		0.10	27.52	9.87

Table 3 shows the effect of formula use on the water absorption capacity of bioplastics. Based on the results obtained, it shows that the average swelling percentage of bioplastic using banana peel flour has the lowest yield and the highest is cassava peel flour. Where the average swelling of bioplastic mango seeds, banana peels, cassava peels and avocado seeds respectively is 32.38%; 30.7%; 34.67%; and 32.36%. The four types of flour used, it is known that banana peel flour has the least starch content compared to other flours, namely 12.78%, while cassava peel, mango seeds and avocado seeds have starch contents of 59%, 58% and 24% respectively. %. This is in line with research Mumtazah et al., (2021), that starch has the ability to absorb water because it has hydroxyl groups. Starch molecules contain very large hydroxyl groups, so their ability to absorb water is also large. The greater the starch content, the greater the hydroxyl groups and their ability to absorb water.

The different plasticizers used give different results, where the average swelling value for the glycerol plasticizer is much higher compared to sorbitol and PVA, namely 35.52% respectively; 30.21%; and 31.84%. According to Isotton et al., (2015), this is due to the higher hygroscopic behavior of glycerol, which interacts strongly with water and is easily incorporated into hydrogen bond chains.

It was found that on average each addition of a greater concentration of beeswax caused a lower swelling percentage, namely 36.02% for 0.04 grams and 28.9% for 0.1 grams. This is because beeswax has hydrophobic properties. Hydrophobic properties are properties where the material has no attraction to water, so that bioplastics that have a high beeswax content are able to easily release water and make the swelling process difficult (Mujahid, 2019).

Water Content According to Treatment Combinations

The water content in bioplastics is an important parameter for producing bioplastics with a plasticizing effect. The results of the water content test in bioplastics can be seen in Table 4, which shows the results of the analysis of water content in bioplastics with a combination of treatments. Based on the selection of starch flour, it can be analyzed that the average water content in the use of mango seed starch flour; banana peel; cassava peel; and avocado seeds each 12.86; 12.30; 13.76; 12.88. From these results, it can be seen that banana peel starch has the lowest water content. This is because banana peels have the lowest starch among the four starches. According to research by Mumtazah et al., (2021), starch has the ability to absorb water because it has hydroxyl groups. The smaller the starch content, the smaller the hydroxyl groups and their ability to absorb water.

Using different plasticizers produces different water contents. Average water content using glycerol plasticizer; sorbitol; and PVA respectively 15.76; 13.85; 9.25. It can be seen that biodegradable plastic using glycerol plasticizer has the highest water content. This is in accordance with research conducted by Valderrama Solano & Rojas de Gante (2014), which states that glycerol attracts more water than other plasticizers.

Obtained from table 4, the water content with the addition of 0.04 grams and 0.10 grams has an average of 13.08 and 12.82. From this average value, the greater the addition of beeswax, the water content contained in biodegradable plastic decreases. This is due to the hydrophobic nature of beeswax so that the water content in biodegradable plastic decreases when the amount of beeswax increases. Like previous research conducted by Rusdi et al., (2022), that materials that have hydrophobic properties will bind less water so that they have a lower water content.

CONCLUSION

In conclusion, differences in the use of starch, type of plasticizer, and amount of beeswax affect its properties when testing is carried out. The four tests carried out include tensile strength and elongation tests, percent degradation tests, percent swelling tests, and water content tests. The conclusions are as follows:

1. The effect of differences in the use of starch in testing tensile strength and elongations was obtained. The higher the starch content used, the lower the tensile strength value, but the opposite is true for the elongation value. In the percent degradation test, it was found that the higher the starch content in the flour, the higher its resistance to degradation. In the percent swelling test, it was found that the higher the starch content, the higher the percent swelling value, as well as in the water content test.
2. The influence of the different types of plasticizer used in tensile strength and elongation testing showed that the PVA type had the greatest tensile strength value while the greatest elongation was using the glycerol type. In the percent degradation test, it was found that the glycerol plasticizer degraded more easily because glycerol more easily absorbs water content, making it easier for microorganisms to degrade. In the percent swelling test, it was found that the glycerol type had the highest value because of the hygroscopic nature of glycerol. A similar thing can be seen in the water content test, which states that the water content in the glycerol type is the highest.
3. The effect of different amounts of beeswax in tensile strength and elongation testing is that the addition of beeswax at low concentrations can increase the tensile strength and elongation values. In the percent degradation test, it wasof a greater concentration of beeswax causes an increase in the duration of the degradation rate. In the percent swelling test, it was found that the addition of a greater concentration of beeswax caused a lower swelling percentage. In testing the water content, it was found that the greater the addition of beeswax, the water content contained in biodegradable plastic decreases.

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