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Biodegradability Study on the Bioplastic Derived from Banana Peel Fruit Waste with Various Ripening Stages

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Bioplastic is currently being used to replace synthetic plastic utilized in food packaging. Bioplastic can be derived from a bio-based product such as banana peel which has a high biodegradation rate. In order to produce bioplastic which can be degraded easily by different types of soil and a good ripening stage must be chosen. The objectives of this research are to evaluate the effect of different types of soil and soil burial time on biodegradability of the films and to analyze the physical appearance of bioplastic derived from various banana ripening stages due to biodegradation process. In this research, bioplastics from the unripe, ripe and overripe peel of *Musa acuminata x balbisiana* (ABB) cv. Awak and *Musa acuminata* (AAA) cv. Berangan were used to analyze the biodegradation rate by means of weight loss. A biodegradability study of the bioplastic produced was conducted and a few variables such as different types of soil which are garden soil with loam (GL) and garden soil with loamy sand (GLS) and soil burial time were evaluated in depth. Moreover, the bioplastic films before and after buried in soil were also analyzed by sensory test and microscopic test. This research shows bioplastic from ripe peel has the highest weight loss (0.0834 g) compared to unripe peel (0.1446 g) and overripe peel (0.2526 g) when buried in GLS due to the increased sugar content in the ripe peel that promotes microbial activity as well as high moisture content and fine texture of the soil. The cross-view of bioplastic film also shows there is a biodegradation process occurred where the microstructure of the bioplastic film is irregular after being buried in the soil.

Keywords*:* Biodegradability; banana peel; bioplastic; soil burial test; fruit waste

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Plastics are widely used in every sector of the industry for instance food packaging, textile, electric and electronic industries. Plastics are meltable and hydrophobic synthetic polymers produced from petroleum such as polyethylene, polypropylene, polyamides, and polyesters Hubbe et al. (2020). Plastics are known for their lightweight, low-cost, and long-lasting qualities (Arikan & Bilgen, 2019). Recent development has led researchers to produce bioplastics created from natural polymeric components such as starch, vegetable oil, cellulose, and lignin, as well as animal-derived compounds like proteins and lipids (Nandiyanto et al., 2020). Production of bioplastic nowadays is typically starch-based which is mainly from corn, rice, or cassava starch with the addition of plasticizer such as glycerol, sorbitol or combination of glycerol and sorbitol to increase the strength and flexibility of the bioplastic produced (Shafqat et al. (2021), Nasution & Wulandari, 2021). Moreover, bio-based bioplastics will tend to increase the biodegradation rate since they are made from renewable resources and easily degraded by the ground.

Bioplastics can be made from the fruit waste with the addition of starch and glycerol as a plasticizer. Fruit waste such as banana peels that contributes 35 – 45% from the total weight of the banana was left unconsumed and thrown away at the landfills (Ganesh et al. (2021) and mostly came from Small and Medium Industries (SMIs) of food such as production of banana fritters, banana chips and traditional desserts (Surattanamal et al., 2022, Yunus, Asman & Mohd. Ali et al., 2020, Tock et al., 2010).

Banana peels consisted of cellulose, starch, pectin, and other polymers that they can be utilized to make bioplastics with fully biodegradable behaviour. However, bioplastics derived from banana peels with various ripening stages (unripe, ripe and overripe) contained different composition of starch and hemicellulose due to some degradative events produced by endogenous enzymes during ripening process. Besides, the level of glucose in banana peels will rise as they get older. In any event, if the peels are overripe, the starch will be converted to glucose, whilst the least ripe peels will be too stiff despite being abundant in starch molecules (Azieyanti et al., 2020). As the ripening stage increased, the sugar content increased which contribute to the perfect environment for the growth of the microorganism. Therefore, microorganism grow rapidly and degraded the bioplastics in the soil. In addition, the biodegradation process is also influenced

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by the environment in which they are found such as type of soil, burial time, pH of soil and temperature of the soil Kumari et al. (2018).

Therefore an attempt has been made to evaluate the relationship between the biodegradation parameters (type of soil and soil burial time) and its biodegradability (in the mean of weight loss) as these behaviour has not been explain in detail in previous studies. Therefore, the objectives of this research are to evaluate the effect of different types of soil and soil burial time on the biodegradability of the films and analyse the physical appearance and microstructure of bioplastic derived from various banana ripening stages due to the biodegradation process.

MATERIALS

Two different types of soil are garden soil with loam (GL) and garden soil with loamy sand (GLS). Unripe, ripe, and overripe of *Musa acuminata* (AAA) cv. Berangan and *Musa acuminata x balbisiana* (ABB) cv. Awak. Sodium hypochlorite solution, 0.5 M hydrochloric acid, 2 mL of 20% glycerol solution, 3 mL of 3% of rice starch, 3 mL of 0.5 M sodium hydroxide solution, and 5 mL of 5% sorbitol were obtained from the laboratory.

The rice starch solution was prepared by dissolution of 3 g of rice starch powder with distilled water until the volume reached 100 mL while the glycerol solution was prepared by dissolution of 20 mL of 100% glycerol with distilled water until the volume reached 100 mL and sorbitol was prepared by dissolution of 5 g sorbitol with distilled water until the volume reached 100 mL. Loam soil was prepared by mixing silt (40%), sand (40%) and clay (20%) while loamy sand was prepared by mixing silt (10%), sand (70%) and clay (20%). Then, the garden soil was mixed with loam soil and loamy sand with a ratio of (3:1) respectively to promote microbial diversity. Moreover, before being utilized to bury the samples, all soil were sieved down to particles smaller than 2 mm in diameter and well mixed. The method used was according to Ghasemlou et al. (2022) and Blunk et al. (2019) with a slight modification.

Preparation of Banana Peels

The unripe banana peels were removed from the flesh by using a stainless-steel knife. Then, about 300 g of unripe banana peels were dipped in sodium hypochlorite solution and the peels were placed in a beaker containing 800 mL of water and boiled for 30 minutes. Next, the water was decanted off and the peels were allowed to dry for 30 minutes at room temperature. Then, the banana peels were placed in a 800 mL beaker. All of these steps were repeated for ripe banana peels and overripe banana peels. The method used was according to Sultan & Johari (2017) with a slight modification.

Preparation of Bioplastic Films

The unripe banana peels were pureed by using a hand blender to form a fine paste. After that, 25 mL of pureed paste was placed in a 100 mL beaker. 3 mL of 0.5 M hydrochloric acid was added and the mixture was stirred by using a glass rod until it is well mixed. Next, 2 mL of 20% glycerol solution and 5 mL of 5% sorbitol were added and mixed well. 3 mL of 3% rice starch which acts as a filler was added and the mixture will be mixed again. Then, 3 mL of 0.5 M sodium hydroxide solution was added to the mixture and mixed well. The mixture was poured onto the baking paper which is on the tray and spread into a thin layer with a glass rod. The tray was then placed in an oven with a temperature of 130℃ for 1 hour and 30 minutes. The bioplastic films were stored in a desiccator to retain the moisture. All of these steps were repeated for ripe banana peels and overripe banana peels. The method used was according to Sultan & Johari (2017) with a slight modification.

CHARACTERIZATION

FTIR Analysis

The FTIR analysis was done with slightly modification from Yaradoddi et al. (2021). The bioplastic sample was subjected to FTIR analysis in range of 4000 cm^{-1} – 650 cm^{-1} using the Fourier Transform Infrared Spectroscopy. The resolution of the spectrum is 1 cm-1 with the scan rate of 4 scans per second in the Attenuated total reflection (ATR) mode.

Effect Different Type of Soil

Two different types of soil which are garden soil with loam (GL) and garden soil with loamy sand (GLS) were used for the soil burial test. For the first step, all of the bioplastic films were weighed before being placed in the soil. Then, all types of soil were placed in plastic cups (12 plastic cups for each soil type). Next, three different ripening stages of bioplastic films which are unripe, ripe and, overripe (2 bioplastic films for each stage) of (1.5 cm x 1.5 cm) were buried in each soil at 6 cm depth then sealed with parafilm and incubated at room temperature for 27 days with sampling time for every day. On the next day, the buried films were cleaned from the soil and weighed then the films were buried again. The method used was according to Nissa et al. (2019) with a slight modification conducted in duplicate.

Effect of Soil Burial Time

By using the same materials and method from the previous test, all the bioplastic films were buried for 27 days with sampling time for every day. For every day of observation, the biodegradation rate through weight loss of the films was analyzed for all types of bioplastic films to determine how many days the

Film need to biodegrade. The method was that used was according to Nissa et al. (2019) with a slight modification conducted in duplicate.

Sensory and Microscopic Test

All the bioplastic films before and after were observed in terms of colour, smell, texture and physical appearance of the bioplastic films were pictured. The method that was used was according to Azmin et al. (2020) with a slight modification. All the bioplastic film before and after bury in soil were observed under a stereo microscope (Rax Vision) with 1.0 magnification. The method that was used was according to Chen et al. (2021) with a slight modification.

RESULT AND DISCUSSION

FTIR Characterization

FTIR spectra in Figure 1 exhibited a large absorption peak ranging from 3,500-3,000 cm^{-1}, which was assigned to the intramolecular hydrogen bonding of hydroxyl groups in water with other constituents in raw banana peels. Apart from that, this peak also attributed to the complex vibrational stretching of hydroxyl groups that existed naturally in carbo-hydrates structures. In addition peak at 2,900 cm^{-1} was attributed to the aliphatic –CH stretching originated from cellulose, hemicellulose and lignin observed in raw banana peels. Besides that, this sharp peak was also attributed to the –CH stretching group originated from starch, amylose and amylopectin existed in raw banana peels. Peaks around 1,600 cm^{-1} is referred to the bending of –OH group in water molecules. Peak around 1400 cm^{-1} is attributed to the –HNCO amide group (amide III) which was originated from protein that can be found in starch structure (Zhang, W., Li, X., & Jiang, W., 2020). Finally, peak at 1000 cm^{-1} is attributed to the –C-OH, C-C and –C=O stretching vibration in polysaccharide groups. Figure 2 showed three major peaks at 3300 cm^{-1}, 1600 cm^{-1} and 1000 cm^{-1} which attributed to the stretching vibration of –OH group originated from starch, stretching vibration of the –C=O bond of Amide I (protein of starch) and –C-O, C-C groups originated from starch (Vijayalaksmi et al., 2022 & Tibolla et al., 2019).

Different Type of Soil

A burial of bioplastic films from various ripening stages was carried out in garden soil with loam soil (GL) and garden soil with loamy sand (GLS). Prior to study, soil moisture content for each soils were recorded and the value were 1.60% (GL) and 1.72% (GLS) respectively.

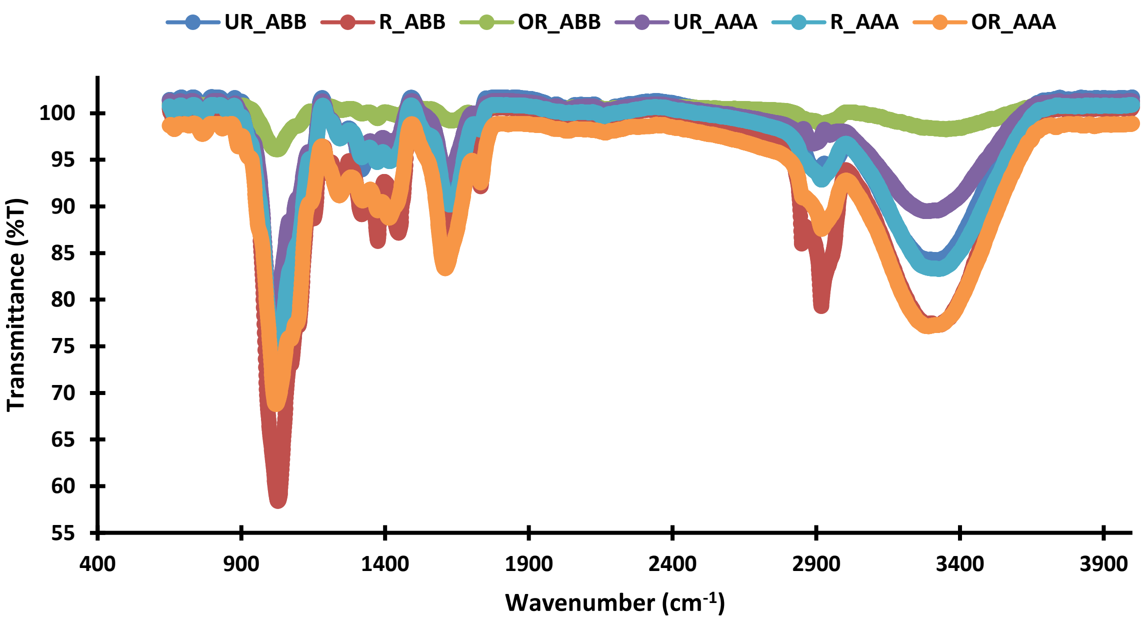

Figure 1. FTIR spectra for raw banana peels of *Musa* ABB cv. Awak & *Musa* AAA cv. Berangan with different ripening stages.

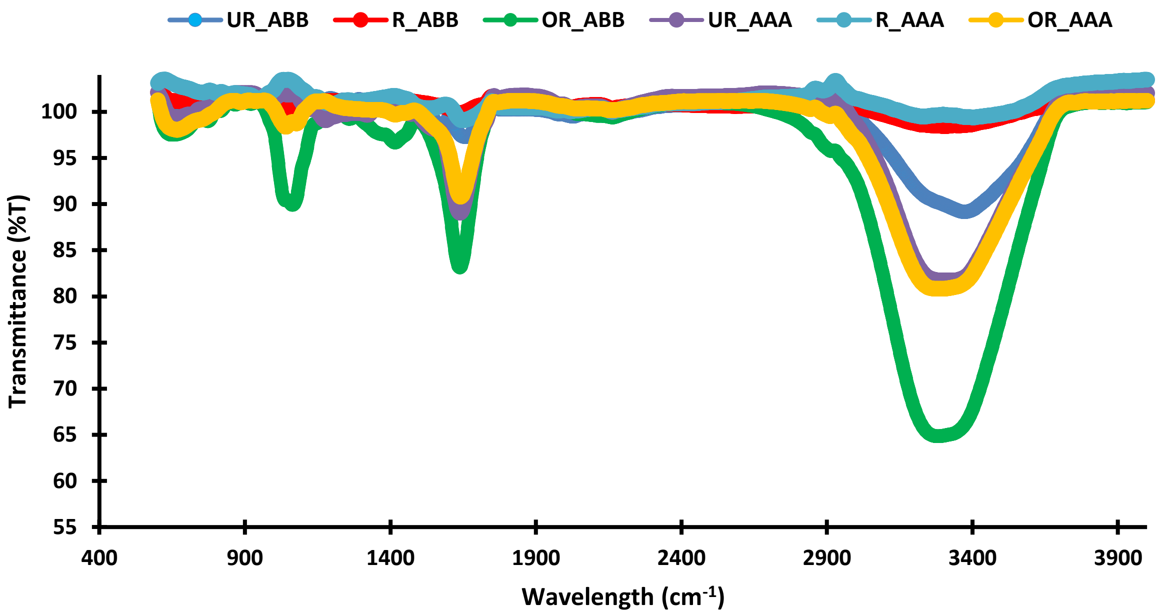

Figure 2. FTIR spectra for bioplastic of *Musa* ABB cv. Awak & *Musa* AAA cv. Berangan with different ripening stages.

The trend of weight loss of bioplastic film derived from peels of *Musa* ABB cv. Awak and *Musa* AAA cv. Berangan were shown in Figure 3. Overall bioplastic derived from various ripening stages showed biodegradability behaviour in both soil type which contain garden soil. Garden soil contains a lot of microbes and minerals, this soil already promotes the biodegradation process of the bioplastic films. Bioplastics of over ripe *Musa ABB cv Awak (OR-ABB)* exhibit highest weight loss for both soil (GL & GLS). Ripe and overripe stage has a high concentration of glucose which leads to high microbial activity when bury in the soil. Thus, the biodegradation rate of banana peel at the overripe stage would be higher compared to unripe and ripe stages. The difference in soil moisture content between GL (1.6%) and GLS (1.72%) affects the biodegradation process in which the moist condition led to the increase in biodegradation efficiency due to the hydrolysis and vigorous activity of microbes Briassoulis & Mistriotis., 2018 and Kumar et al., 2022.

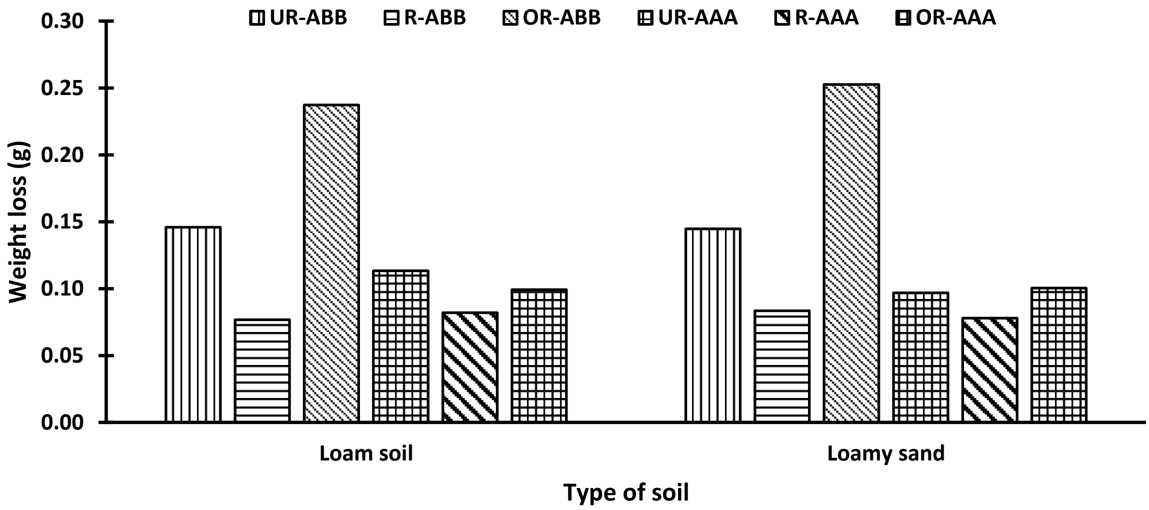

Figure 3. Weight loss of bioplastic film derived from *Musa ABB* and *Musa AAA.*

Effect of Soil Burial Time

In this study, 27 days were allocated to observe the weight loss of bioplastic films buried in GL and GLS. The actual weight of bioplastic films was recorded before burying in the soil. It was observed that on the first day of burial, the weight of the bioplastic films derived from peels of *Musa* ABB cv. Awak and *Musa* AAA cv. Berangan was increased when buried in both types of soil. The increased weight of the bioplastic films may be due to the absorption of moisture from the soil into the bioplastic film since soil contains water and bioplastic film produced with the combination of glycerol and sorbitol has made the bioplastic to absorb moisture (Shafqat et al., 2021). However, the weight of the bioplastic film decreased gradually on day two and followed by the next day.

As shown in Figures 4 and 5, the weight loss of bioplastic films from unripe, ripe and overripe peels of *Musa* ABB cv. Awak and *Musa* AAA cv. Berangan were decreased when buried in GL and GLS. This proved that bioplastic film derived from bio-based was successfully degraded in soil. A study made by Rohmawati et al. (2018) shows that after being buried in the soil-compost mixture for 30 days, the bioplastic decreased in mass on average by 44.12%. This demonstrated that bioplastics could be broken down more quickly than non-biodegradable ordinary plastics. Additionally, in comparison to the weight loss of bioplastic film from unripe and ripe peels, the bioplastic film from ripe peels shows a great weight loss compared to unripe peels. This demonstrates that the biodegradation process has occurred vigorously in bioplastic film from the ripe peel. According to Schulz et al. (2021), in the later stages of edible life, fruits have the highest amounts of fructose and glucose. This is why the weight loss of bioplastic film from the ripe peels is higher than the unripe and overripe peels. Besides, the weight loss of bioplastic film from the unripe peel is greater compared to the overripe peel due to the addition of plasticizer that made the texture of the unripe bioplastic film became moist compared to the bioplastic film from ripe and overripe peels. The moisture from the bioplastic film from the unripe peel encourages the microorganism to biodegrade the film better than the bioplastic film from the overripe peel.

A recent study by Schulz et al. (2021) shows that sugar content increase as the ripening stage increase thus will enhance the biodegradation rate. However, in this study, the result shows a slightly contradiction at the ripening stage of ripe and overripe in which the weight loss of the bioplastic film from the overripe peels is lower which also shows low biodegradation rate compared to the bioplastic film from unripe and ripe peels. Research made by Watharkar et al. (2020) shows that at the overripe stage of *Musa nana Lour* banana pulp, there was a decrease in total sugar (glucose and fructose), which may have been brought on by a weaker starch hydrolysis reaction, a rise in moisture, and a fall in total soluble solids. Moreover, a study conducted by Phillips et al. (2021) shows that the total sugars in overripe banana is lower compared to ripe banana. Thus, this explains why the biodegradation activity and weight loss of bioplastic film from overripe peel are lower as the total sugar increase from unripe to ripe then decrease from ripe to overripe. Besides, depending on the species and growing conditions, such as relative humidity, temperature, and soil, the concentration of glucose, fructose, and sucrose can change (Yap et al., 2017).

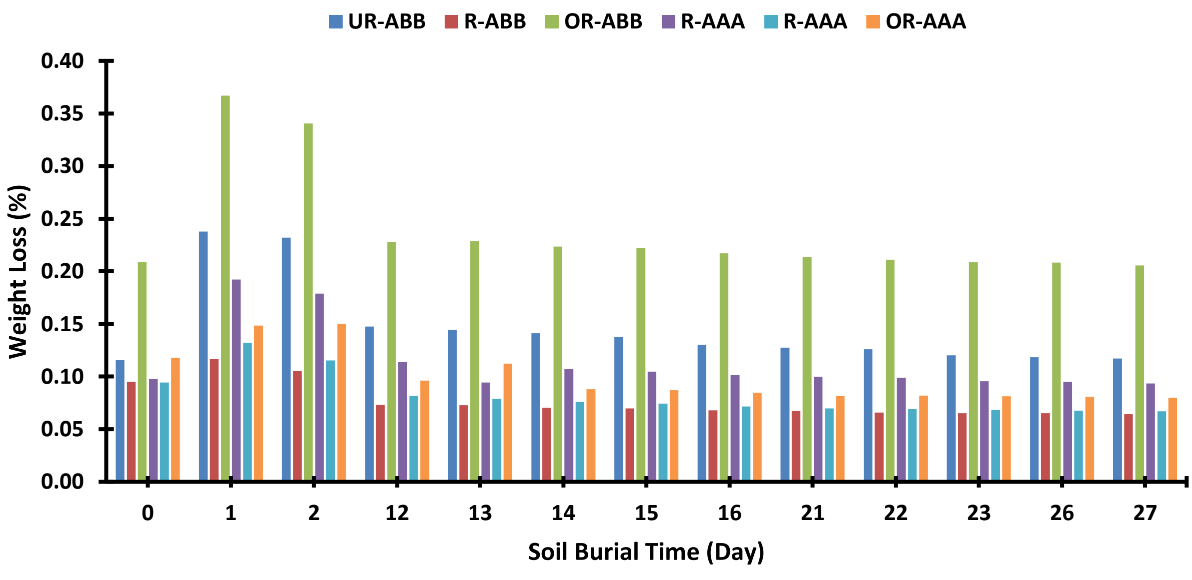

Figure 4. Weight loss of bioplastic derived from *Musa* ABB cv. Awak and Musa AAA cv. Berangan versus soil burial time, loam (GL).

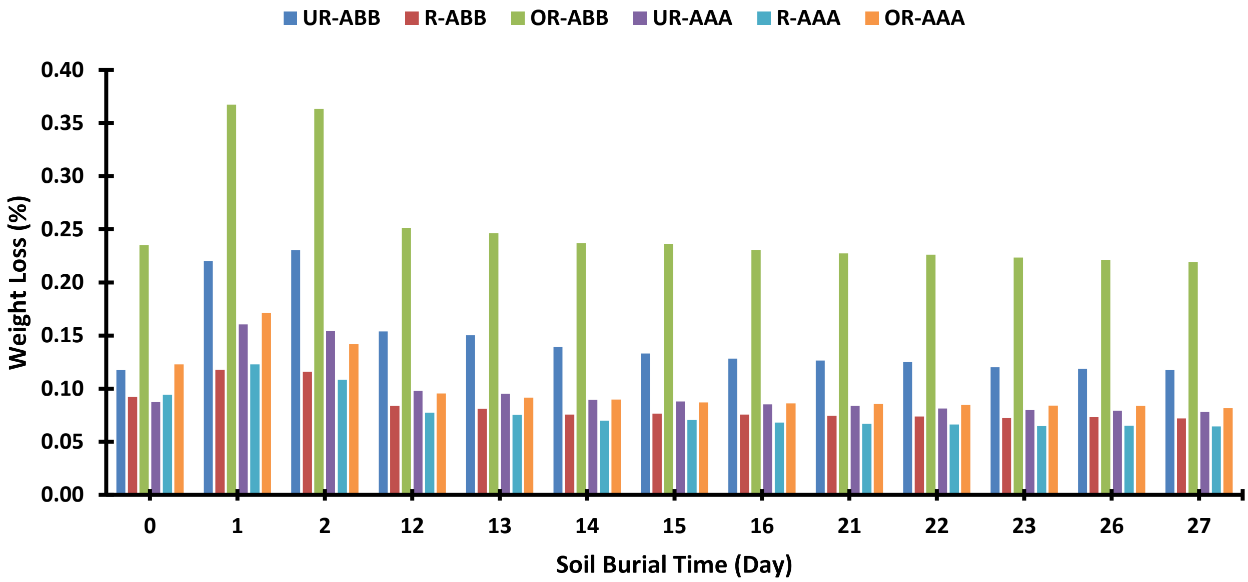

Figure 5. Weight loss of bioplastic derived from *Musa* ABB cv. Awak and Musa AAA cv. Berangan versus soil burial time, loamy sand (GLS).

Bioplastic derived from peel of *Musa* ABB cv. Awak shows a great biodegradation rate in which high weight loss has obtained compared to bioplastic peel. *Musa* AAA cv. Berangan. Moreover, the peel of *Musa* ABB cv. Awak take a short time to convert from green to yellow compared to *Musa* AAA cv. Berangan. This is because as the peel become matured, the sugar content increased while the chlorophyll content become decreased thus increase the bio-degradation rate of the bioplastic film. A study made by Wu et al. (2019) shows that at 30°C, yellowing was increased in *Musa* ABB cv. Awak while green ripening was only seen in the *Musa* AAA cv. Cavendish fruit. It was determined that the green ripening of AAA fruit was particularly caused by the fruit's inhibited chlorophyll breakdown has occurred vigorously in bioplastic film from the ripe peel. According to Schulz et al. (2021), in the later stages of edible life, fruits have the highest amounts of fructose and glucose. This is why the weight loss of bioplastic film from the ripe peels is higher than the unripe and overripe peels. Besides, the weight loss of bioplastic film from the unripe peel is greater compared to the overripe peel due to the addition of plasticizer that made the texture of the unripe bioplastic film became moist compared to the bioplastic film from ripe and overripe peels. The moisture from the bioplastic film from the unripe peel encourages the microorganism to biodegrade the film better than the bioplastic film from the overripe peel.

Sensory and Microscopic Evaluation of Films

Table 1 (before buried in soil) shows the sensory and microscopic evaluation of bioplastic film according to the colour, texture, smell, physical appearance and microstructure. As shown in Table 1, the appearance of bioplastic film from unripe, ripe and overripe peels derived from *Musa* ABB cv. Awak and *Musa* AAA cv demonstrates the colour changes started from brownish to black. A study made by Azmin et al. (2020) shows the colour of the bioplastic is more brownish when it has the maximum cellulose concentration by which the concentrations of cellulose to fiber were 0:100, 25:75, 50:50, 75:25 and 100:0. Moreover, as looking at their texture, each gives a different texture. Overripe bioplastic film derived from *Musa* ABB cv. Awak and *Musa* AAA cv. Berangan had a dry and hard texture. In addition, a study made by Azmin et al. (2020) shows that bioplastic film from cocoa pod husk shows a decrease in cellulose percentage making the bioplastic film becomes drier and tougher.

Besides, unripe and ripe bioplastic film had a sour smell while overripe bioplastic film had a sweet smell. A research made by Phillips et al. (2021) shows the mean fiber in unripe, ripe and overripe fruit was 18 g, 4-5 g and 2 g respectively and research by Azmin et al. (2020) shows the increase of fiber at certain concentration led to sweet smell but turns to odourless when the fiber is at the Thus, this explains why the biodegradation activity and weight loss of bioplastic film from overripe peel are lower as the total sugar increase from unripe to ripe then decrease from ripe to overripe. Besides, depending on the species and growing conditions, such as relative humidity, temperature, and soil, the concentration of glucose, fructose, and sucrose can change (Yap et al., 2017).

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After bioplastic films were buried in GL and GLS as shown in Table 2, all colour of the bioplastic films become darker due to the interaction and biodegradation process in the soil. Moreover, all the bioplastic becomes dry and rough as the moisture from the bioplastic film decreased throughout the burying time. Nevertheless, this research shows smell of the bioplastic film had slightly changed and some remained. The smell observed might be due to the existence of cellulose and fiber content in the banana peel.

The stereo microscope images show the actual cross-sectional view of the bioplastic film which can be concluded that these bioplastic films had uneven surfaces and holes that can be seen clearly. However, the cross-sectional view of the bioplastic film after burying was not shown any obvious changes but the colour of the bioplastic film was changed after burying in the soil. Moreover, Rohmawati et al. (2018) stated that due to the amorphous nature of the polymer, this bioplastic was easily destroyed, allowing soil-dwelling bacteria to assault the bioplastic molecules. This is why the cross-view of the bioplastic film after burying shows a little scattered compared to the bioplastic film before burying in the soil.

CONCLUSION

Bioplastic films derived from the peel of various ripening stages of *Musa* ABB cv. Awak and *Musa* AAA cv. Berangan shows there is biodegradation activity occurred when buried in GL and GLS. Bioplastic film derived from the peel of ripe shows a great biodegradation activity in which this bioplastic film has a higher weight loss when buried in GL and GLS compared to bioplastic film derived from the peel of unripe and overripe of the bananas. This is followed by high weight loss of bioplastic film from the peel of unripe then overripe. Bioplastic film from the ripe peel has the highest weight loss due to the increase sugar content that promotes the biodegradation activity by the bacteria to become vigorous.

Table 1. Sensory and microscopic evaluation of bioplastic films for *Musa* ABB cv. Awak and *Musa* AAA cv. Berangan before soil burial test.

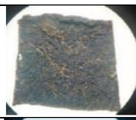
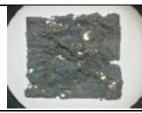
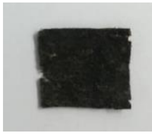
Banana ripening stages	Sensory evaluation colour	Texture	Smell	Physical appearance (before buried)	Microscopic images (before buried)
UR-ABB	Slightly brownish	Slightly moist, brittle, rough	Sour smell		
R-ABB	Golden brown	Moist soft	Slightly sour		
OR-ABB	Black	Dry, rough, hard	Slightly sweet		
UR-AAA	Brownish	Slightly moist, slightly rough,	Sour smell		
R-AAA	Dark brown	Dry, slightly soft	Sour smell		
OR-AAA	Dark brown	Dry, slightly rough, hard	Slightly sweet		

Table 2. Sensory and microscopic evaluation of bioplastic films for *Musa* ABB cv. Awak and *Musa* AAA cv. Berangan after soil burial test.

Type of soil	Banana ripening stages	Sensory evaluation colour	Texture	Smell	Physical appearance (after buried)	Microscopic images (after buried)
GL	UR-ABB	Dark brown	Dry, brittle, rough	Slightly sour		
GLS						
GL	R-ABB	Dark brown	Dry, brittle	Slightly sour		
GLS						
GL	OR-ABB	Black	Dry, rough, hard	Slightly sweet		
GLS						
GL	UR-AAA	Dark brown	Dry, rough, brittle	Slightly sour		
GLS						
GL	R-AAA	Black	Dry, rough, brittle	Slightly sour		
GLS						
GL	OR-AAA	Dark brown	Dry, rough, brittle	Slightly sweet		
GLS						

Nevertheless, bioplastic film from overripe peel has the lowest weight loss may be due to the reduction of sugar content as the ripening stage is increased by weak hydrolysis of starch. Thus, low biodegradation activity has occurred when burying in GL and GLS. Thus, bioplastic film derived from ripe peel is better than unripe and overripe peel. Furthermore, bioplastic film buried in GLS gives a positive weight loss due to the high moisture content and fine texture of the soil that served the best condition for the biodegradation process of the bioplastic film compared to GL. Moreover, each bioplastic film gives a different texture, colour, smell and physical appearance. It shows that bioplastic film from unripe peel has light brown colour and is slightly moist compared to bioplastic film from ripe and overripe peel. The cross-view of each bioplastic film observed by using a stereo microscope shows that biodegradation activity has occurred since there is an irregular pattern and cross-link of the bioplastic film before and after being buried in GL and GLS.

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