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Effect of Polyvinyl Alcohol on Cassava and Potato Starch Plastic Film: Mechanical, Thermal and Swelling Properties

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Abstract: Uncontrolled use of conventional plastic has triggered environmental concerns due to its non-biodegradable properties. Due to these circumstances, biodegradable plastic made up from natural polymers like cassava and potato starch was developed. However, due to the brittleness and low mechanical strength of starch-based bioplastic film, it has contributed to the development of polymer blending between starch and polyvinyl alcohol (PVA) to produce starch/PVA plastic film. The addition of glycerol as a plasticizer in this starch bioplastic films formulation also able in reducing the brittleness of the bioplastic films and preventing the tearing during the peeling process. By using the casting method, various proportions (30% and 60%) of 4% PVA solution were added into the starch film-forming solution and the effect of PVA content to cassava and potato starch bioplastic films was investigated. It was discovered that by increasing the PVA percentages, the thickness of both cassava and potato starch bioplastic films were reduced, while the moisture content, swelling capacity, and biodegradation rate were increased. The bioplastic films from cassava starch showed a higher swelling capacity and biodegradation rate compared to potato starch due to its increment in hydrophilicity after blending with PVA. It was also discovered that the tensile stress for CP60 and tensile strain for PP30 increased significantly. In FTIR analysis, all samples showed C-H stretching of starch and PVA, and OH group and C-O-C stretching that could enhance the properties of bioplastic films from the formation of a hydrogen bonding network. The Tg of CP30 decreased with the increase of PVA content due to the increment of hydrophilicity of bioplastic films.

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Therefore, adding PVA in cassava and potato starch films can be used as water soluble bioplastic for bettering future waste management.

Keywords: Bioplastic, Cassava Starch, Potato Starch, Polyvinyl Alcohol

1. Introduction

Plastic has become something inseparable to human daily life and has been used indispensably in various applications such as packaging, automotive, electronics, aerospace and medical. One of the applications of plastic where it is used tremendously in our daily life particularly at every home is for food packaging. Conventionally, plastic was made up from polymeric materials which is non-biodegradable. This type of material is used to act as a protection or cover from bacterial infection from surrounding to the food as well as for preservation by keeping the shelf life of the food longer.

This immense use of plastic leads to other consequential effect such as higher production of solid waste due to its undegradable properties. Currently, the proper methods to dispose this kind of material are recycling, disposal, landfill, and incineration [1]. In addition, improper plastic waste disposal like littering will end up in terrestrial and marine ecosystems that give adverse impact to wildlife and alter the fertility of soil due to its toxicity. The continuation of this current trend will lead to 12,000 million metric tons of plastic waste on Earth by 2050 [2].

Due to these circumstances, the production of alternative degradable plastic packaging has been studied further by using renewable or natural-based sources such as starch. Starch is available abundantly from many types of plants such as cassava, potato, corn, wheat, banana etc. Thus, many studies had led to developing an environmental-friendly plastic film that is not just cost efficient, but also helps in creating a sustainable environment [3,4,5,6,15, 20]. Starch also can be used as a natural polymer due to its tendency for biological degradation by living microorganisms. It has a high intensity in absorbing moisture content due to the presence of hydroxyl group. However, challenges arisen while developing this kind of starch-based film is the brittleness and low mechanical property which hindered the use of this kind of material commercially.

Because of these challenges, modification of starch bioplastic films with polyvinyl alcohol (PVA) is necessary. PVA is a synthetic polymer that is also biodegradable and environmentally friendly. Previous studies have shown that PVA-based plastic film has high transparency and tensile properties, adjustable water solubility and non-toxic bioplastic films [3]. However, due to its relatively high cost, the addition of polymer blending was done to enhance the mechanical property of the starch-based bioplastic films while at the same time reducing the cost of developing degradable plastic materials [20].

As the casting method is used in this study, the amount of water and temperature during the gelatinization are key factors that need to be controlled as there is no shear stress and pressure applied [21]. During the starch gelatinization process, the presence of water causes the starch granules to swell and lead to the solubilization of amylose and amylopectin molecules. This makes the water molecules diffuse into the starch polymers chains and make the starch gelatinize and form the starch-based bioplastic films [22].

Comparison study was made based on the properties of starch-based bioplastic films from cassava and potato starch that were blended with the different proportion of PVA which were 0%, 30% and 60% by undergoing some physical characteristic test, thermal, swelling capacity and biodegradable analysis.

2. Materials and Methods

The cassava starch (Cap Kapal ABC) and the potato starch (Cap Bintang) were purchased from Thye Huat Chan Sdn Bhd, Malaysia, polyvinyl alcohol was purchased from Eva Chem, Malaysia and glycerol anhydrous from R&M Chemicals, Malaysia.

2.1 Bioplastic Preparation

In this study, the casting method was used as it is a laboratory-scale production and presents the excellent quality of bioplastic films. This method involved the effect of starch's solubility in a solvent under heat with a temperature above gelatinization temperature. This is to induce the starch gelatinization process and produce the gelatinized and solubilized solution. The solvent casting method was carried out to prepare the biodegradable plastic films with different composition of 4% PVA solution as according to the study by Gómez-Aldapa et al. [22] which is 0, 30 and 60%. The composition of samples with different proportions of PVA can be referred in Table 1. The preparation of this biodegradable plastic films was referred to the method by Sharmila et al. [4] with some modifications. The filmmaking procedure was started by heating 200ml of distilled water until it reached 40^{o}C. Once it reached 40^{o}C, 12g of cassava starch was poured into the heated distilled water while stirring continuously. Then, glycerol which acted as a plasticizer in this plastic film preparation was added in the ratio 1:3 (glycerol: starch (dry basis)). The mixture was stirred continuously for 30 minutes while the temperature was increased gradually from 40^{o}C to 80^{o}C (±5^{o}C). During the pregelatinized starch at a temperature 50^{o}C, 4% PVA solution was added in different proportions (30% and 60% of starch weight). The stirring of mixture was continued at 80^{o}C (±5^{o}C) for another 30 minutes to ensure a complete homogenization. The solution became viscous and thick. Continuous stirring was important to keep and maintain the temperature of the film-forming solution. During the stirring process, there would be many air bubbles trapped in the mixture. Because of that, 30ml of the solution was poured into the centrifuge tube and undergoing centrifugation at 1000rpm for 3 minutes. This can help to reduce the existence of bubbles in bioplastic films. After that, 30ml of the mixture was cast into the petri dish with diameter 9cm and dried in the laboratory oven (Memmert, UF160, Germany) at a temperature 50^{o}C for 18 hours. Once dried, the bioplastic films were peeled off the petri dish, kept in the air-tight plastic bag and stored in the desiccator before characterization analysis. This procedure was repeated by using potato starch. All the obtained bioplastic films were named with CP0, CP30, CP60, PP0, PP30, and PP60 indicating the percentage of 4% PVA composition to each sample. The samples were left to dry for at least 48 hours prior to the characterization.

Table 1: The composition of samples with different proportion of PVA

Samples	Proportion of PVA (%)	Constituents			
		Starch (g)		Glycerol (g)	PVA (g)
PP0	0	Potato	12	3	0
PP30	30	Potato	12	3	3.6
PP60	60	Potato	12	3	7.2
CP0	0	Cassava	12	3	0
CP30	30	Cassava	12	3	3.6
CP60	60	Cassava	12	3	7.2

2.2 Bioplastic Analysis

Upon fabricated, the samples were undergone several analysis and characterization in term of the morphology, thickness, moisture content, mechanical property, and swelling capacity. Later, the samples were subjected to characterization using Fourier Transform Infrared Spectroscopy (FTIR) to investigate the effect of organic bond in the samples, thermal property to investigate the effect of high temperature onto the samples and last but not least biodegradability test to investigate the effect of

degradability of the samples in soil. All samples were subjected to statistical analysis where samples with significant difference values of less than 0.05 (p<0.05) [30].

2.2.1 Thickness

The thickness for all bioplastic samples were measured using micrometer with precision 0.001mm (Mitutoyo, M110-25, Japan). Measurements were carried out in 5 (five) random points on each sample [24]. The results were expressed as average value by using the Eq. (1).

$$thickness = \frac{sum\ of\ measured\ values}{5} \qquad Eq.1$$

2.2.2 Moisture Content

Plastic film was cut into 2x2 cm. The initial weight of bioplastic film was weighed. The sample was dried for 24 hours at 105ºC and the final weight was weighed [24]. The moisture content was determined by the Eq. (2). The test was carried out in triplicate to calculate the mean of moisture content for each sample.

$$Moisture\ content\ \% = (initial\ weight - final\ weight)/(initial\ weight)\ x\ 100 \qquad Eq.2$$

2.2.3 Mechanical property

The tensile strength for each sample were measured by using texture analyser (TA. XT Plus, Stable Micro Systems Texture Technologies, Scardale, NY, USA). The program used was Tortilla Extensibility with test speed 2.00 mm/sec and operated at room temperature, with 1.00 mm/sec crosshead speed. Firstly, samples of plastic film were cut into 7x2 cm. The tensile stress and tensile strain were determined by using Eq. (3) and (4) with original cross-sectional area 5x2 cm. The test was carried out in triplicate to calculate the mean of both tensile strength for each sample.

$$Tensile\ stress\ (MPa) = Force\ (N)/(Initial\ cross\ sectional\ area\ (m^2)) \qquad Eq.3$$

$$Tensile\ strain = Extensibility\ (m)/(Original\ length)m \qquad Eq.4$$

2.2.4 Swelling Capacity

Each sample that was cut into 2x2 cm were weighed and recorded the initial weight. After that, the weighed bioplastic films were immersed in the test tube that contained 20ml of distilled water for 1 hour as there were samples that started to dissolve if further immersion was done. After 1 hour, the samples were carefully taken out and the excess or attached water from the samples surface were dried carefully by tissue paper. After that, each immersed samples were weighed again to know the final weight and calculated the percentage of swelling capacity (SC) by using Eq (5). The test was carried out in triplicate to calculate the mean percentage of SC for each sample [25].

$$SC\ (\%) = (Final\ weight\ (g) - initial\ weight\ (g))/(Initial\ weight\ (g))\ x\ 100 \qquad Eq.5$$

2.2.5 Fourier Transform Infrared Spectroscopy (FTIR)

The functional group for all bioplastic films with different concentrations of 4% PVA solution were analyzed by using Fourier Transform Infrared Spectroscopy (Perkin Elmier 99365 Spectrophotometer, USA). This analysis obtained the information regarding the interaction between PVA and starch for both cassava and potato. By using sample that was cut into 1x1 cm, the analysis was carried at room temperature where wavenumbers ranged from 700 to 4000cm-1 [4, 29].

2.2.6 Thermal Property

The thermal properties for all samples were analyzed by using Differential Scanning Calorimetry (DSC) (Shimadzu, DSC-60Plus, Japan). Each bioplastic film was cut into very tiny pieces and 5 to

10mg of each sample were placed in an aluminum pan which then was sealed. An empty pan was used as reference. Bioplastic films samples were heated from 20 to 200ºC under constant heating rate at 20ºC/min and held for 3 mins, according to method by Tian *et al*. [9]. After that, the cooling scan was conducted from 200-20ºC at 20ºC/min cooling rate.

2.2.7 Biodegradability

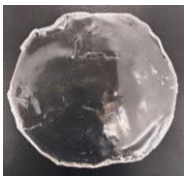
A compost soil was filled into a rectangular perforated plastic container with tiny holes at the bottom and each side of containers. Each bioplastic films (2x2cm) were weighed to determine the initial weight before burying deep to 5 cm and kept at ambient temperature (outside laboratory, atmospheric surrounding without direct sunlight). It was sprayed with 10ml of distilled water once a day to keep the soil moist. After 7 days, the sample were carefully unburied and washed with distilled water to get rid the excess of dirt. After that, the washed samples were dried for 18 hours at 50ºC and cooled in a desiccator prior weighed to determine the final weight. The degradation rate was calculated by using Eq. (6). The test was carried out in triplicate to calculate the mean of biodegradability. Meanwhile, for the samples that was disabled to wash to get rid of the dirt, it was photographed to observe the disintegration visually.

$$Biodegradation\ rate\ (\%) = \frac{Initial\ weight\ (g) - Final\ weight\ (g)}{Initial\ weight\ (g)}\ x\ 100 \qquad Eq.6$$

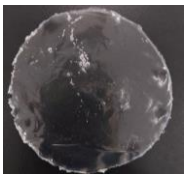
3. Results and Discussion

3.1 Film appearance

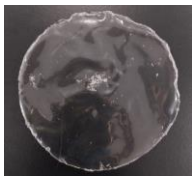
The morphology of cassava and potato starch bioplastic films is depicted as in Figure 1. All samples were discovered showing homogenous and smooth surface of bioplastic films, while samples CP0, CP30, and CP60 appeared to be stickier than PP0, PP30, and PP60, and becoming less sticky with the increment of PVA content. All samples were also discovered to be easy to unmold when blending with 4% PVA solution. According to Gómez-Aldapa *et al*. [5] a good compatibility in producing homogeneous formulations can be observed during processing and developed samples. This polymer blending proves that homogenous thick solutions after gelatinizing of starch were successfully formed. However, it also can be seen from Figure 1, there is a visible formation of air bubbles on potato starch bioplastic films which might happen when air is trapped naturally between the folds of high viscosity film forming solutions during stirring and casting process. Another study by Tarique *et al*. [6] had reported the similar phenomenon where potato starch bioplastic films tend to have more bubbles than cassava starch bioplastic film. This phenomenon was due to the viscosity of film forming solutions that were made up from potato was thicker than cassava starch mixture and increased after being mixed with PVA during the gelatinization process.



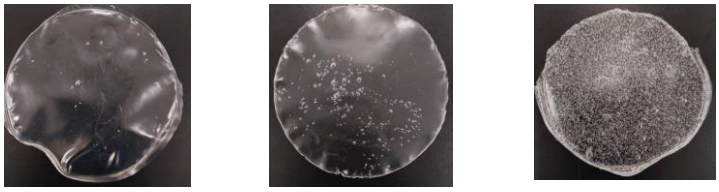
(CP0)



(CP30)



(CP60)



(PP0) (PP30) (PP60)

Figure 1: The physical appearance of cassava and potato starch bioplastic films with different proportion of PVA

3.2 Thickness, moisture content and swelling capacity

The thickness, moisture content and swelling capacity of both starch bioplastic films with different proportions of PVA are depicted in Table 2. Both cassava and potato starch bioplastic films display the similar effect in term of thickness where, as the concentration of PVA increased, the thickness of bioplastic films decreased. This finding is supported from previous study by Aguirre-Loredo et al. [7], which stated that the thickness of bioplastic films is affected by the presence of linear polymer chain where it shows that the bioplastic films with higher amylose content (linear structure) obtaining the smaller thickness of bioplastic film. Thus, when the formulation of starch bioplastic films mixed with PVA, it increases the number of linear polymer chains presence [5] and cause the reduction of bioplastic films thickness if the concentration of PVA increases. However, there is no significant different (p<0.05) between the cassava and potato starch bioplastic film regarding the effect of PVA to the thickness of film.

From the moisture content analysis in Table 2, it indicates that starch bioplastic films showed an increment of moisture content when mixed with PVA, while potato starch bioplastic films did not show a linear experimental data like cassava starch. This phenomenon happened due to the increment of OH group in starch which increases the hydrophilicity of starch bioplastic films [8]. However, there is no significant different (p<0.05) between potato and starch bioplastic films regarding the effect of PVA to moisture content of bioplastic films.

On the other hand, based on Table 2 also, PP30, PP60, CP30, CP60 samples showed higher swelling capacity when mixed with PVA content. This is due to the present of the OH group in both cassava and starch plastic film which increased when concentration of PVA increased during the polymer blending, which led to increment of swelling to the bioplastic films. This phenomenon was also obtained by Tian et al. [9] where in the study reported, the increase of starch content in starch: PVA ratio, reduces the water uptake of bioplastic films which indicates that PVA possessed higher swelling capacity than starch. Besides, another study by Gómez-Aldapa *et al*. [5] also states that the polymer blending between PVA, and potato starch causes the bioplastic films to be more hydrophilic due to increase of OH group. It can be clearly seen that cassava starch plastic film has higher swelling capacity than potato starch plastic film. According to another previous study by Vamadevan & Bertoft [10], the study shows that the higher of amylose content in starch, the lower the swelling of granules. Due to that, by referring to El Seoud *et al*. [11], that states potato starch has higher percentage of amylose content than cassava starch which is 21% and 16.7%, respectively, has shown that the high existence of amylose content in potato starch has retards the swelling capacity of bioplastic film during the water immersion. During the analysis, the bioplastic films from cassava starch bioplastic films almost dissolved. Thus, cassava starch bioplastic film with high existence of amylopectin content has more intensity of water uptake as it contributes to the swelling property by maintaining the integrity of starch granules until it collapses [12].

Table 2: Thickness, moisture content and swelling capacity of cassava and potato starch bioplastic films with different proportion of PVA

Samples	Thickness (μm)	Moisture content (%)	Swelling capacity (%)
PP0	0.308 ± 0.06	22 ± 1.34	88.02 ± 3.2
PP30	0.288 ± 0.05	24.15 ± 7.09	112.73 ± 1.67
PP60	0.270 ± 0.04	23.75 ± 5.94	211.58 ± 9.16
CP0	0.276 ± 0.06	18.35 ± 3.87	481.08 ± 4.12
CP30	0.252 ± 0.03	22.14 ± 2.58	536.41 ± 11.95
CP60	0.246 ± 0.07	24.22 ± 7.77	661.68 ± 14.28

± standard deviation (n=3 except thickness n=5)

3.3 Mechanical property

The tensile stress and tensile strain of cassava and potato starch bioplastic films with different proportions of PVA are depicted in Table 3. The data for tensile stress in this study is slightly lower than the other reported works [4,22] that might be due to the low concentration and weight of PVA used in the formulation. Table 3 shows that the tensile stress for CP60 increases significantly when PVA was added into the formulation which indicates that the intermolecular interactions between PVA and cassava starch increase [13,19]. This phenomenon can also be seen in the study by Gómez-Aldapa et al. [5] where the tensile stress of bioplastic films with the proportion of PVA higher than starch, increase significantly when the PVA content is more than 40%. According to Majdzadeh-Ardakani & Nazari, [14] which research about the corn starch/PVA/clay nanoparticles, had stated that enough PVA content could enhance the tensile property of bioplastic films. Meanwhile, the tensile strain for PP30 shows a significant increase when PVA was added. The increased in tensile strain of potato starch bioplastic films with PVA blending due to the interruption of starch crystallinity that happens from the formation of hydrogen bond between the OH group from PVA and interference of hydrogen bonds among starch granules [15].

Table 3: The tensile strength and tensile strain of cassava and potato starch bioplastic films with different proportion of PVA

Sample	Tensile stress (N/m^2)	Tensile strain (%)
PP0	24784.23	21.96
PP30	20188.1	32.89
PP60	23948.63	35.97
CP0	15928.33	40.52
CP30	16295.93	38.94
CP60	40427.80	41.24

3.4 FTIR analysis

Figure 2 shows the FTIR spectra used to observe the interaction that could occur between starch and PVA. It showed that the interaction of starch with PVA is not significantly visible during FTIR analysis. However, it shows that all samples with different proportions of PVA shared the same

characteristics of broad band ranging from 3264 to 3268cm^{-1} that shows the frequencies of hydroxyl groups of PVA and starch in the bioplastic films [16, 28]. All the bioplastic samples show that there is presence of C-H stretching ranging on 2926 to 2928cm^{-1}. There is a sharp peak on 1149cm^{-1} for C-O vibration of C-O-C groups that indicates the glucose unit in starch [17]. According to Patil et al. [17], the high number of OH and CO groups can enhance the compatibility between starch and PVA content by forming hydrogen bonding network that improves the properties of bioplastic films. Table 4 shows the summary for existing functional group in each bioplastic sample.

Table 4: The identified functional group of cassava and potato starch bioplastic films with different proportion of PVA

Wavenumber (cm^{-}1)	Band assignment
3000-3300	OH stretching
2926	C-H stretching of CH_2
1149	C-O vibration

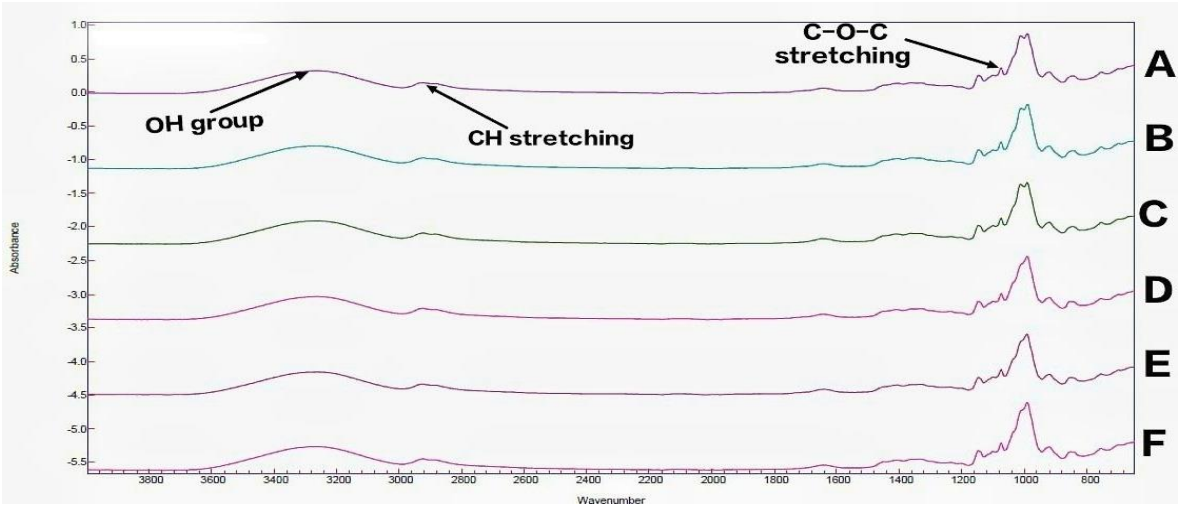


Figure 2: The overlaid spectra for cassava and potato starch bioplastic films with different proportion of PVA (A: CP30, B: CP60, C: CP0, D: PP30, E: PP60, F: CP0)

3.5 Thermal Property

Table 5 shows the glass transition temperature (T_g) for samples of cassava and potato starch bioplastic films with 0%, 30% and 60% of PVA proportion in their formulation. However, the experimental data is lower than the previous study [23]. In this study, CP30 shows a significant decrease of T_g after blending with PVA content. As the increment of PVA causes the increase of bioplastics hydrophilicity, the T_g also decreases due to the water absorption which leads to plasticizing effect [26]. PVA is also considered as a plasticizer that interrupts the interaction of hydrogen bonds among starch molecules which cause the films to be softer and flexible [16].

Table 5: The T_g of cassava and potato starch bioplastic films with different proportion of PVA

Samples	T_g (°C)
PP0	28.84
PP30	29.43
PP60	27.73
CP0	36.55
CP30	28.17
CP60	28.01

3.6 Biodegradability

Based on Table 6, it was discovered that the addition of PVA into the potato starch bioplastic films increased the degradation rate. Study by Ismail & Zaaba [18], also reported the same experimental data where the higher the proportion of PVA in the starch bioplastic films, the higher the biodegradation rate. During the observation, only potato starch bioplastic films were weighed while cassava starch bioplastic films were observed visually which can be seen from Figure 3. The biodegradability of bioplastic films from cassava starch is higher than potato starch. The increment of swelling capacity and hydrophilicity of bioplastic films has induced the biodegradation rate by favoring the growth of microorganisms and bacteria to break down the biopolymers. Thus, the biodegradation rate of both starch bioplastic films increased, as the PVA content increased. However, cassava starch bioplastic films had higher biodegradability than potato starch bioplastic films as the bioplastic films started to disintegrate with soil adhered over the cassava starch bioplastic films.

Table 6: The biodegradation rate of potato starch bioplastic films with different proportion of PVA after 7 days of soil burial test

Samples	Biodegradation rate (%)
PP0	31.78 ± 1.36
PP30	35.13 ± 1.60
PP60	39.72 ± 2.10

± standard deviation (n=3)

(CP0) (CP30) (CP60)

Figure 3: The observation of cassava starch bioplastic films with different PVA content after 7 days of soil burial test

4. Conclusion

Cassava starch and potato starch bioplastic films were prepared by blending with various proportions (30% and 60%) of 4% PVA solution and undergone casting. Thickness, moisture content, swelling capacity, tensile strength, FTIR and biodegradability test had been observed and determined. The increment of PVA in starch bioplastic films had reduced the thickness and increased the moisture content of both cassava and potato starch bioplastic films with no significant different (p<0.05). This polymer blending had increased the swelling capacity and biodegradation rate, while bioplastic films from cassava starch shows a higher swelling capacity and biodegradation rate than potato starch plastic film. The tensile strength of CP60 and tensile strain of PP30 show a significant increase after blending with PVA. In FTIR analysis, there was the existence of C-H stretching that are present in PVA and starch, and existence of OH group and C-O-C stretching that could enhance the properties of bioplastic films by forming hydrogen bonding network. Besides that, the T_g of starch bioplastic decreased with the increased of PVA content due to increment of hydrophilicity of bioplastic films. Thus, adding PVA in cassava and potato starch bioplastic films can be used as soluble bioplastic for the betterment of future waste management.

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