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## FABRICATION AND CHARACTERIZATION OF JACKFRUIT SEED POWDER AND POLYVINYL ALCOHOL BLEND AS BIODEGRADABLE PLASTIC

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### **ABSTRACT**

The biodegradable plastics (BGP) were fabricated from the blend of jackfruit seed powder starch and poly(vinyl alcohol) (PVA). Development of BGP is highly recommended and getting extensive interest to develop an environmentally friendly material that can replace the petro-chemical plastics. The aim of this research is to fabricate the BGP from the jackfruit seeds/PVA blend and characterize the mechanical and biodegradability properties. Results on the mechanical properties of the fabricated BGP shows that by increasing the composition of jackfruit seed powder starch, the tensile strength of the BGP was significantly decreased. The biodegradability properties of the samples were determined by monitoring the structure of fabricated BGP by using Fourier Transform Infrared Spectroscopy (FT-IR). The FT-IR results indicated that the structural properties were changed after 8 weeks of soil burial test by comparing with control samples. BGP with high amount of jackfruit seed powder starch shows better biodegradability characteristics.

**Keywords**: Jackfruit seed; poly(vinyl alcohol); mechanical properties; FTIR spectroscopy; biodegradable plastic

### **INTRODUCTION**

Petroleum based plastics commonly used for packaging, construction, electronics, furniture and machinery because of its desirable convenience, versatility and low cost. Based on the survey in 2001, there are roughly 230 million tons of waste which are not recyclable was produced in United States and nearly 11% of the waste is plastic [1]. Sandra and Matthew (2017) reported that a million plastic bottles are bought every minutes and will increase to another 20% by 2021, thus could create an environmental crisis [2]. Currently, the common way to dispose all the petroleum based plastics waste is by incineration produces emission of several greenhouse gasses such as carbon dioxide. Moreover, the abundance of petro-chemical based polymers that take ages to discompose also results in landfilling. A lot of effort has been taken to reduce the usage of the petroleum based plastics such as recycling and replacing plastics with another environmental friendly material such as paper products. Michael (2016) reported that only 2% of the world's 300 million tons of plastic production comes from plant [3]. Therefore, researchers need to find an alternative way to replace the petroleum based plastics. Currently biodegradable plastics (BGP) are an increasingly well-known alternative to petroleum-based plastics and have been highlighted owing to their inherent biocompatibility.

One of the main raw materials that have been used by researchers to produce the BGP is starch. Starch is one of the universal raw material that is available all over the world. Besides that, starch is a completely biodegradable polysaccharide produced by a large number of plants and also inexpensive material [4]. One of the major drawback associated with the use of starch for the fabricating BGP is its poor mechanical properties [4]. Furthermore, packaging films fabricated from entirely of starch will affect the strength and rigidity to withstand the stress, hence limit the application [5].

Poly(vinyl alcohol) (PVA) is one of the most popular water-soluble polymers that have been used in packaging applications [6]. According to Ishigaki et al. (1999), disadvantage of using PVA in producing plastics is having a lower biodegradability compared to other biodegradable plastics which approximately take 300 days to degrade about 75 % of its weight [7]. However, the blend of starch with PVA seems to be an alternative way to overcome the slow rate of the biodegradability of the PVA because of the enzyme and bacterial activity in the BGP [7]. Besides, PVA itself has substantial tensile strength, better flexibility, and hardness together with gas and aroma barrier characteristics which are expected to further improve its physical and mechanical properties. Azahari et al., 2011 reported from his previous study on PVA/corn starch blend that the Scanning Electron Microscopy (SEM) micrograph shows the corn starch granules dispersed well in the 70/30 PVA matrix which helps to improve the mechanical properties of the film tested and agreed with the tensile property results [5].

Inspired by those studies, the aim of this work is to fabricate the BGP from the jackfruit seeds/PVA blend and characterize the mechanical and biodegradability properties.

## **EXPERIMENTAL**

All the chemicals used in this study were of analytical grade and used without further purification. Deionized (DI) water was used throughout this experiment. The jackfruit seeds used in this experiment were fresh and purchased from local market. The jackfruit seed arils and spermoderms were peeled and cleaned until only white seed flesh remains. Following that, the seeds were chopped into small pieces and dried using an oven at temperature of 70 °C for 24 hours. Then, the dried jackfruits were ground and sieved to obtain seed flour particles of diameter 56 um.

PVA solution was prepared by dissolving the PVA into the distilled water. The mixture solution was heated at 100 °C for 90 minutes. On the other part, the jackfruit seed flour was mixed with 0.1 M hydrochloric acid and dissolved in distilled water. The mixture was then stirred at 80 °C for 10 minutes until the flour is completely dissolved. While stirring, 0.1 M sodium hydroxide (NaOH) solution and glycerol were added into the jackfruit seed mixture solution to neutralize the mixture.

After that, the jackfruit seed mixtures were poured into PVA solution and continuously stirred at 100 °C for 30 minutes until the homogenous gel-like mixture was formed. In this study, different composition of PVA/ jackfruit seed blend was used as represented in Table 1. Finally, the mixture was casted by using casting plate with size 20 cm x 20 cm and dried in an oven at 45 °C for another 24 hours.

Mechanical properties of each samples were measured by using Vantage Thwing-Alber Horizontal Tensile Tester machine. Three replicates of each samples were cut according to the American Standard Method (ASTM) D882 for the material with thickness less than 1.0 mm. The length and width of the measured samples were cut into dimension of 50 mm times 15 mm, respectively. The reading of the measurements was recorded and analyzed. The biodegradability was followed by Fourier Transform Infrared Spectroscopy (Perkin Elmer).

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**Table 1** Material Compositions

Sample	PVA (%)	Jackfruit Seed Powder (%)
100/0	100	0
70/30	70	30
50/50	50	50
30/70	30	70
0/100	0	100

### RESULTS AND DISCUSSION

#### **Mechanical Properties**

The mechanical properties of the samples in term of tensile strength, percentage of elongation of break and the Young's modulus were analyzed. Fig.1 shows the effect of PVA/Jackfruit Seed Powder Ratio on the Percentage of Elongation at Break. The major component of Jackfruit seed powder is starch which constitutes approximately 78% the total Biomass [8]. It can be observed that, increasing the amount of the jackfruit seed starch in the blend decreases the tensile strength of the BDG. This might be due to the increasing amount amorphous nature of jackfruit seed starch that leads to a lower tensile strength of the BGP. P. Naknaen et al., (2017) found from the XRD diffraction pattern that degree of crystallinity of the native jackfruit seed starch was 29.6%. The lower degree of crystallinity showed the higher degree amorphous area results [9]. Pure PVA of ratio 100/0 in the fabricated BDG has the highest tensile strength compared to other samples with value of 11.25 MPa. The tensile strength continues to decrease with the ratio of 70/30, 50/50 and 30/70 with the tensile strength of 2.97 MPa, 1.24 MPa and 0.60 MPa, respectively. The sample with ratio 0/100 cannot be prepared for mechanical properties measurement due to high difficulties to mold into desired size and because of having high brittleness properties. PVA having higher numbers of hydroxyl group and this properties allows it to react with many types of functional groups, hence lead to higher strength and better mechanical properties [10-11]. Normally, starch cross-linking is performed by treating starches (semi-dry or slurry) with reagents capable of forming either ether or ester linkages between hydroxyl (-OH) groups on starch molecules [12]. The results from this work show a similar trend with previous study on PVA/corn starch blend conducted by Azahari et al., (2011). He reported that, the decrease in tensile strength with the incorporation of the starch is due to the filler-matrix interaction [5]. Higher starch loading makes the filler-filler interactions become more pronounced compared to filler-matrix interactions [5]. Similar behavior was also observed by Kormin et al., (2017) that the higher composition starch in LDPE led to decrease the mechanical properties because of the starch incorporated into LDPE still retained their granular shape after processing [13].

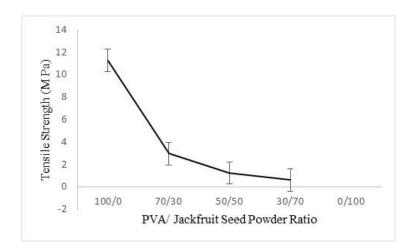


Fig. 1 Effect PVA/Jackfruit Seed Powder Ratio on the tensile strength

Fig. 2 shows the percentage of elongation at break for different composition of PVA and Jackfruit seed powder ratio. The result shows the decreasing percentage of elongation at break by increasing the jackfruit seed powder in the blend. It can be stipulated that this result is due to the amorphous nature of jackfruit seed starch that led to the brittleness properties. It can be observed that the brittleness property of the PVA/Jackfruit seed powder contributes to the decrease of the elongation at break percentage which also agreed by Sadhu *et al.* [10]. Its due to incorporation of the starch where the filler-matrix interaction whereas with higher starch loading, filler-filler interactions become more pronounced than filler-matrix interactions [5].

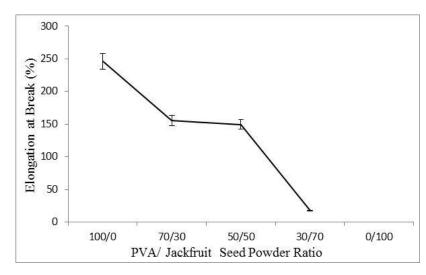


Fig. 2 Effect PVA/Jackfruit Seed Powder Ratio on the Percentage of Elongation at Break

## Biodegradability

The biodegradability properties of the fabricated BGP were determined by using Perkin Elmer Spectrum FTIR. Each sample was cut into dimension of 5 mm<sup>2</sup>. Then, the samples were buried in the top soil (pH 6.5) from Unit of Field, Universiti Teknologi Mara Pahang at a depth of about 5 cm. The test was performed according the method published by Obasi *et al.*, (2013) whereas the top soil was placed into plastic containers with tiny holes perforated at the bottom and on the side of the container to increase air and water circulation [14].

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The soil was kept moist with water and stored at room temperature 30 °C throughout the test period. The soil burial test lasted for eight weeks. After that, the samples were taken out, washed with distilled water, dried in and oven at 40°C and tested by using the FTIR spectrometer. The obtained spectra from FTIR spectrometer were analyzed to identify the biodegradability of the sample. FTIR was used to analyze the compound structures and the functional group of materials. Fig.3 illustrates FTIR spectra of PVA/ Jackfruit seed starch blend at different compositions. In comparison of all spectra generated, all samples show consistency in peaks and these peaks corresponding to their respective functional groups of monomer units in polymeric chains. The strong sharp band frequency at 3600 cm<sup>-1</sup>  $^{1}$  can be attributed to the stretching and bending vibrations of the functional hydroxyl group [15-16]. This result shows that the increasing jackfruit seed starch in the blend leads to the decreasing peak height. This might be due to the hydrogen bonding becomes weaker in PVA/jackfruit starch blend than in pure PVA because of the diminution in the number functional hydroxyl group [17]. The reduction of the peak height may result from the interaction of different -OH groups in the starch and PVA molecular chains [18]. This indicates that all of the hydroxyl groups in the PVA molecular chain and starch are involved in the blending process. The peak around 2200 cm<sup>-1</sup> indicate  $C \equiv C$  stretch bond with alkynes functional group. The band around 1710 cm<sup>-1</sup> shows that the frequency is corresponding to carboxyl and ester carbonyl bands, - C=O- stretching [19]. The spectra peak intensity of PVA/ Jackfruit seed starch blend increased with increasing the percentage of jackfruit seed starch in the blend. This result showed similarity with previous reports in literature whereas they found that the peak intensity increased for the starch/polyvinyl alcohol/citric acid ternary blend 3:3:0.08 compared with the starch/polyvinyl alcohol/citric acid ternary blend 1:1:0 film [20]. The vibrational peaks are assigned to O–H stretching, C–H stretching, and  $C \equiv C$  stretching and they existed in the FTIR spectra of PVA/starch blends indicating the success of blending of PVA with starch [21].

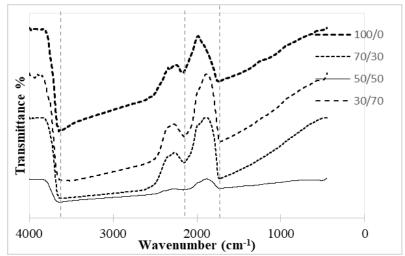


Fig. 3 FTIR spectra for different ratio of PVA/Jackfruit seed starch blend

For this analysis, samples with PVA/Jackfruit seed starch ratio 100/0, 70/30 and 70/30 were buried in the soil for 8 weeks to observe the degradability of the samples. The FTIR results of the buried samples were compared with the control samples. Fig. 4 (a), (b) and (c) showed the comparison spectra for the control samples and after eight weeks of soil burial test. Fig. 4(a) showed there was a slight change in terms of peak position as well peak intensity after soil burial test for eight weeks at region (I) 3700 – 3500 cm<sup>-1</sup>, (II) 2700  $-2300 \text{ cm}^{-1}$ , (III)  $2200 - 2000 \text{ cm}^{-1}$  and (IV)  $1800 - 1600 \text{ cm}^{-1}$ . Meanwhile Fig. 4(b) and (c) can be seen that, there are major changes in terms of peak position and peak intensity for both types of samples at region (I) 3700 – 3500 cm<sup>-1</sup>, (II) 2700 – 2300 cm<sup>-2</sup>  $^{1}$ , (III) 2200 - 2000 cm  $^{-1}$  and (IV) 1800 - 1600 cm  $^{-1}$ . At region (I), the reduction of peak intensity occurred after eight weeks soil burial. This region was assigned to the -OH- stretching bond. However, compared to sample 30/70 ratio, higher reduction of peak intensity –OH- stretching bond was observed than sample 70/30 ratio. Region (II) in the range of 2700 – 2300 cm<sup>-1</sup> showed the peak position were shifted to from 2510 cm<sup>-1</sup> to 2630 cm<sup>-1</sup> for composition 100/0, 2515 cm<sup>-1</sup> to 2720 cm<sup>-1</sup> for composition 70/30, and 2515 cm<sup>-1</sup> to 2520 cm<sup>-1</sup> for composition 30/70 with reduction of peak intensity –C-H- bond and increasing of -OH- starching bond after eight weeks for burial test. Region (III) in the range of  $2200 - 2000 \text{ cm}^{-1}$  is a characteristic of  $-C \equiv C$ - stretching bond existed on the control samples and the peak intensity was reduced after eight weeks for soil burial test. Similar observation also found in the spectra for region (IV) in the range of 1800 – 1600 cm<sup>-1</sup> due to -C=O- stretching bond become lesser after eight weeks of soil burial. By comparing the changes in the infrared spectra, it can be found that sample 30/70 ratio of PVA/ Jackfruit seed starch blend exhibited a significant change in peak intensity due to the changes of molecular structures compared to other samples. The decreasing peak intensity is relative to the timing of soil burial compared to the initial control spectra [22]. The peak shifted might be due to the microorganism activity during soil burial test. The chemical changes such as shortening of peaks on FTIR spectra also confirmed the degradability of the polymer [23]. These results suggest that higher jackfruit seed starch portion in the blend lead to higher degradability. The study conducted by Ali et al., on the biodegradation of cellulose blended polyvinyl chloride films found that the changes on width of the peak at wavelength 2359 cm<sup>-1</sup> (corresponding to O-H and C-O-C bond stretches) was sharp in control, which was broad in sample attributed by degradation process [23]. Similar behavior was also observed by Azahari et al., for the biodegradability of PVA/starch films whereas the biodegradability rate of the films also increased with increasing the starch content and burial time. Azahari et al., found that biodegradability rate of PVA/starch blends was increased with increasing the starch content when compared with that of pure PVA to reach a maximum weight loss at the starch content of 70%. In the soil, the maximum weight loss was 85% after 8 weeks for films containing 70% starch [5].

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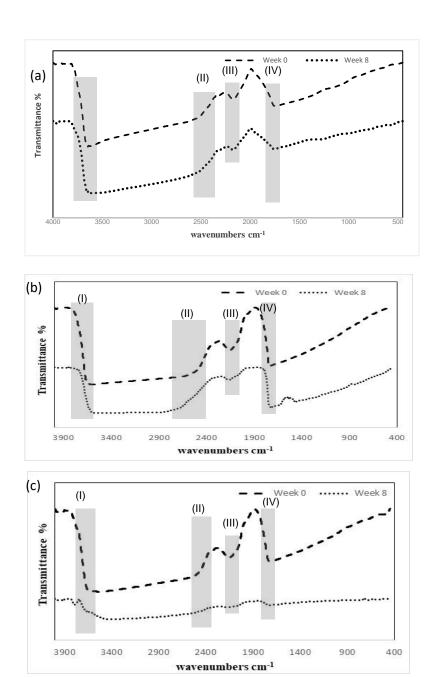


Fig. 4 FTIR spectra for (a) 100/0, (b) 70/30 and (b) 30/70 ratio of PVA/Jackfruit seed starch blend

### **CONCLUSION**

As a conclusion, the biodegradable plastic from PVA/jackfruit seed starch blend was successfully fabricated and characterized for mechanical and degradability properties. The results clearly indicate that, the mechanical properties of the biodegradable plastic from PVA/jackfruit seed starch were improved with increasing the portion of PVA in the blend. Furthermore, the degradability properties were increased and show a significant change in the FTIR spectra as increasing the portion of jackfruit seed starch in the blend. In the future study, it is recommended to improve the fabrication of the samples by adding binder in the fabrication process of biodegradable plastic and can be considered as an alternative to the petroleum-based plastic.

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