Effects of Banana Skin Powder on Properties of Jackfruit Seed Starch/Poly(Vinyl Alcohol) PVA Film

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ABSTRACT. Biodegradable jackfruit seed starch (JFSS)/poly (vinyl alcohol) (PVA) films reinforced with different amount of banana fruit skin powder (BSP) were produced via solution casting method. This study aims to investigate the effect of BSP on the properties of JFSS/PVA films. The blend ratio between JFSS and PVA was fixed at 4:3 wt.% while the composition of BSP was varied from 1 to 2.5 wt.%. The mechanical and physical properties of the composite films were investigated. From the analysis, the optimum blend ratio of JFSS/PVA is achieved at maximum tensile strength 10.90 MPa with 1 wt.% BSP. The results of mechanical properties were supported by scanning electron microscope (SEM) in which the banana skin powder is properly wetted by the JFSS/PVA blend, thus indicates the good interaction within the blend. The reinforcement of BSP into JFSS/PVA blend films has also resulted in the existence of hydrogen bonding interaction evidenced by Fourier transform infrared spectroscopy (FTIR) spectra and increase the percent weight loss of the film over time through soil burial test, hence, proved its biodegradability.

Keywords: Starch/PVA blend, Banana reinforcement, Biodegradable, Composite films;

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1. INTRODUCTION

One third of the plastics produced today are used for packing purposes which will be quickly discarded. The plastics will be accumulated in landfills and causes threat to the environment [1]. From the environment point of view, biodegradable materials have been rapidly developed in the past years. PVA is one of the biodegradable synthetic polymers commonly used, but, its degradation rate is slow. As an alternative to reduce the plastic waste and accelerate the degradation process, PVA frequently blended with other natural polymers to improve its biodegradability. The natural polymer such as starch has high potential in enhancing PVA biodegradability by blending both components. Starch is renewable, inexpensive, biodegradable and abundantly available especially from agriculture field [2]. The usage of starch extracted from agriculture wastes such as jackfruit seed is quite promising, since it does not interrupt food chain and may add value to the starch as well [3]. The blend of starch extracted from fruit seeds and PVA is assumed able to increase biodegradation rate of composite film formed. Adding starch to PVA, however, may reduce its original mechanical properties, hence, the film is reinforced with fruit skin powder as filler to further improve its properties. Currently, natural fibers are gaining so much attention to act as filler or reinforcement in a matrix material because they are eco-friendly, low in cost, abundantly available resources and renewable. In this study, banana skin is utilized as it is considered as one of agricultural waste. Having said that, this natural fiber can be easily obtained from banana skin as the reinforcement for composite material in order to improve its mechanical properties [4].
Therefore, the main objective of this study is to develop biodegradable composite film of poly (vinyl alcohol) (PVA) blended with jackfruit seed starch (JFSS) and reinforced with banana fruit skin powder (BSP) for packaging applications via solution casting method. The relationship between the amount of reinforced fruit skin powder with starch/PVA in composite film on mechanical and physical properties was evaluated. Finally, biodegradability studies through soil burial test were performed to investigate the effect of incorporating reinforcement of BSP in the films degradation.

2. EXPERIMENTAL METHOD

2.1 Preparation of Starch/PVA Reinforced Banana Skin Composite Film. The starch/PVA blend films were prepared via solution casting method proposed [5] with minor alteration. The ratio of starch to PVA was fixed at 4:3 wt.%, while the amount of BSP were varied from 1 to 2.5 wt.% with total mass of the system is 100 g. First, 3 wt.% PVA was dissolved in distilled water was heated up to 95 °C. Then, 4 wt.% starch was added to the mixture and stirred continuously at about 70 °C to ensure the solution is homogeneous. Finally, the films solution was casted over an acrylic plate and oven dried at 35 °C for 24 hours. The films were peeled off and stored in the desiccator maintained at 23 °C and 30% relative humidity prior to characterization. The steps were repeated by adding different weight percent of banana skin powder. Table 1 illustrates the ratio of PVA, jackfruit seed starch and banana skin powder in sample preparation.

Table 1 The ratio of jackfruit seed starch, poly (vinyl alcohol) and banana skin powder

<table>
<thead>
<tr>
<th>Materials</th>
<th>Samples (weight proportions, g)</th>
<th>Jackfruit seed starch</th>
<th>PVA</th>
<th>Banana skin</th>
<th>Distilled water</th>
</tr>
</thead>
<tbody>
<tr>
<td>PVA</td>
<td></td>
<td>0</td>
<td>3</td>
<td>-</td>
<td>97.0</td>
</tr>
<tr>
<td>S-PVA</td>
<td></td>
<td>4</td>
<td>3</td>
<td>-</td>
<td>93.0</td>
</tr>
<tr>
<td>S-PVA-BSP1</td>
<td></td>
<td>4</td>
<td>3</td>
<td>1.0</td>
<td>92.0</td>
</tr>
<tr>
<td>S-PVA-BSP1.5</td>
<td></td>
<td>4</td>
<td>3</td>
<td>1.5</td>
<td>91.5</td>
</tr>
<tr>
<td>S-PVA-BSP2</td>
<td></td>
<td>4</td>
<td>3</td>
<td>2.0</td>
<td>91.0</td>
</tr>
<tr>
<td>S-PVA-BSP2.5</td>
<td></td>
<td>4</td>
<td>3</td>
<td>2.5</td>
<td>90.5</td>
</tr>
</tbody>
</table>

2.2 Tensile Test. The mechanical properties of the composite films such as tensile strength and percent of elongation were evaluated using universal testing machine (Shimadzu; Material Testing System) based on ASTM Standard D882-02 at 20 mm/min of speed under load of 5 kN. Samples of composite film with dimension of 70 mm × 10 mm were cut according to ISO-527.

2.3 Scanning Electron Microscope (SEM) Test. The surface structures of the JFSS, BSP, starch/PVA film with and without reinforcement of BSP were observed using a scanning electron microscope (JEOL, Model JSM 5600 SEM; JEOL Ltd., Tokyo, Japan). The samples were sputter coated with carbon (Polaron SC515) and observed under the SEM at a voltage of 5 kV and 10 kV.

2.4 Fourier Transform Infrared Spectrometry (FTIR) test. Fourier transform infra-red (FTIR) spectra of the BSP, PVA film, starch/PVA film with and without reinforcement of BSP were recorded on FTIR spectrometer (Spectrum 100 FTIR; Perkin Elmer, United States). Measurements were obtained between 4000 cm⁻¹ to 600 cm⁻¹ at a resolution of 4 cm⁻¹ with average of 32 scans per sample.

2.5 Soil Burial Test. Biodegradability of the starch/PVA blend films with and without reinforcement of BSP were investigated through soil burial method [6] with slight modifications. Samples were cut to dimension of 20 mm × 20 mm and the initial mass are recorded. Then, the samples covered with plastic net and placed in
pots containing soil for 20 days with exposure to natural weathering. Each sample are taken for measurement at interval of 5 days for 20 days, dried and weighed on the electronic mass balance. The weight loss of the sample over time was calculated using Eq. 1.

\[
\text{Weight loss (\%)} = \frac{w_i - w_d}{w_i} \times 1
\]  

(1)

where \(w_i\) is the initial weight of the sample and \(w_d\) is the weight of the sample after 5 days.

3. RESULTS AND DISCUSSION

The addition of BSP as reinforcement greatly affect the tensile strength (TS) and elongation at break (E%) of starch/PVA film. Fig. 1 shows the effect of increasing BSP on mechanical properties of starch/PVA films.

![Fig. 1 Effect of increasing banana skin powder content on tensile properties of starch/PVA films](image)

Reinforcement of starch/PVA films with 1 wt.% BSP improves both TS and E%. The TS of film increased by more than 4% which is from 10.45 MPa to 10.90 MPa. At the same time, the E% significantly increased by 59% with addition of 1 wt.% BSP which is from 2.28% to 3.63%. This is because the banana fiber gives strong adhesion between the phases in starch/PVA, thus, provides better intermolecular interactions between starch molecules and fiber or fiber and PVA [5]. However, further addition of BSP more than 1 wt.% deteriorated both TS and E% of starch/PVA film. This is probably due to the agglomeration of the banana fiber, thus, reduce the attachment between the fibers and the starch/PVA that causes by the reduction of wettability of the fiber [7]. The results obtained are supported by morphological structure observed under SEM. The morphology of JFSS granules and BSP (magnification of 1000x), PVA, starch/PVA and reinforced BPS starch/PVA films (magnification of 100x) were presented in Fig. 2. Fig. 2(a) revealed the starch granules have varied round and irregular shape with smooth surface [8]. Meanwhile, Fig. 2(b) revealed the BSP has long entangled structure of fiber with rough surface which is expected to provide strong adhesion when reinforced to starch/PVA blend [9]. Fig. 2(c) shows morphology of unfilled PVA film is smooth and
transparent without any addition of starch. As a result from blending starch with PVA, Fig. 2(d) demonstrates good interactions between starch and PVA. The blend is well dispersed, less agglomerate and no formation of voids occur. Fig. 2(e) illustrates the addition of 1 wt.% BSP result in properly wetted fiber with starch/PVA film, hence gives good interaction between the fiber and starch/PVA. Adding more than 1 wt.% BSP promotes formation of voids and agglomeration of the granules as shown in Fig. 2(f-g). When 2.5 wt.% of BSP is added, formation of crack can be observed. This is due to more fiber for film reaction and bigger void is formed due to pull out from agglomerated BSP as shown in Fig. 2(h).

![Fig. 2 SEM micrographs of (a) jackfruit seed starch, (b) banana skin fiber, (c) PVA, (d) S-PVA, (e) S-PVA-BSP1, (f) S-PVA-BSP1.5, (g) S-PVA-BSP2 and (h) S-PVA-BSP2.5](image)

The FTIR spectra of BSP, PVA, S-PVA and S-PVA-BSP1 are shown in Fig. 3(a-d), respectively. Fig. 3(a) shows the broad peak at 3274.02 cm⁻¹ correspond to O-H stretching related to intramolecular hydrogen bonds of cellulose while the small peak at 2917.83 cm⁻¹ represents C-H bond stretching in cellulose and hemicellulose of the banana skin. The peak of 1732.21 cm⁻¹ and 1012.06 cm⁻¹ indicate the C=O stretching and O-H bond, respectively. The results of FTIR spectrum obtained for BSP is in agreement with the study reported [9] which confirmed that banana skin is one of the cellulosic fibers.

Furthermore, all spectra in Fig. 3(b-d) exhibit the characteristic absorption bands of pure PVA which are 3274, 2919, 1732 and 1243 and 1087 cm⁻¹ which correspond to O-H stretching, C-H stretching, C=O stretching, and C-H wagging of PVA, respectively. These peaks also existed in the FTIR spectra of starch/PVA
blends shown in Fig. 3(c), indicating the success of blending of PVA with starch. Fig. 3(b,c) show the peak intensity reduce with incorporation of starch in PVA. The peak was shifted from 1732.21 to 1715.94 cm\(^{-1}\) and from 1087.95 to 1019.88 cm\(^{-1}\) which means that the C-O in C-O-H group of starch and C=O stretching was involved in the hydrogen bond formation, respectively. This can be concluded that the FTIR analysis for starch/PVA blend showed that more stable hydrogen bond can be formed.

Furthermore, incorporation of banana fiber reinforcement in starch/PVA blend in Fig. 3(d) shifted the characteristic peak from 1715.94 to 1732.52 cm\(^{-1}\) was wider and less intense with incorporation of starch and banana fiber. This indicated that there is O-H that is more easily vibrate, hence more O-H forming hydrogen bond. The change in peak intensity at 3274 cm\(^{-1}\) confirmed the number of the hydroxyl groups rises due the interaction of fiber with starch/PVA blend \[10\]. It is believed that the formation of hydrogen bond based on the FTIR spectra is correlated with the mechanical properties and biodegradability rate of film.

The biodegradability behavior of non-reinforced and reinforced starch/PVA loss and macroscopic appearance shown in Fig. 4(a,b), respectively. The degradation behavior of the films is dependent on the presence of banana fiber reinforced in the starch/PVA blend. The graph shows that, starch/PVA blend has the highest biodegradability rate than reinforced starch/PVA blend films. The blend films incur the highest weight loss at 27.42% probably due to the hydrophilic nature of starch itself. Hydrophilicity increase water absorbability, hence, increase degradation. It is believed that the white spot on the surface of the samples are the microorganisms since starch is their source of food, hence, causing the biodegradability behavior of the samples. In fact, the degradability also can be related with its appearance which is diminished in size which contributed to the weight loss of the samples.Moreover, addition of BSP reinforcement improved properties of the starch/PVA blend film as a whole. Both banana skin and starch are hydrophilic and polar. Incorporation of both components is believed to resist water due to formation of hydrogen bonding between starch and banana skin fiber evidenced by FTIR analysis. Thus, better hydrogen bonding reduced susceptibility of blend film to water \[10\]. It can be concluded that the weight loss of the starch/PVA films at 20 days proved that they are biodegradable.
Fig. 4 Biodegradation of non-reinforced and reinforced starch/PVA films (a) weight loss, and (b) macroscopic appearance after buried in soil for 20 days

4. SUMMARY

This study has shown that banana skin powder effectively reinforced jackfruit seed starch/PVA blend film. Reinforcement of 1 wt.% banana skin powder has achieved the maximum strength of 10.90 MPa. The morphological structure of the film revealed the banana skin powder is well distributed within the blend by the appearance of peak indicating hydrogen bond. In addition, the composite films showed their degradability in soil environment, but considerably decreased the weight loss as reinforcement is incorporated.

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