Preparation and characterization of jackfruit seed starch/poly (vinyl alcohol) (PVA) blend film

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Abstract. From the environmental 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 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. The usage of starch extracted from agriculture wastes such as jackfruit seed is quite promising. In this study, jackfruit seed starch (JFSS)/poly (vinyl alcohol) (PVA) blend films were prepared using the solution casting method. The effect of starch content on the mechanical (tensile strength and elongation to break %) and physical properties of the tested films were investigated. The optimum tensile strength was obtained at 10.45 MPa when 4 wt.% of starch added to the blend. But, decreasing trend of tensile strength was found upon increasing the amount of starch beyond 4 wt. % in starch/PVA blend films. Nevertheless, elongation at break decreases with the increase in starch content. The mechanical properties of the blend films are supported by the Field Emission Scanning Electron Microscopy (FESEM), in which the native JFSS granules are wetted by PVA continuous phase with good dispersion and less agglomeration. The incorporation of JFSS in PVA has also resulted in the appearance of hydrogen bond peak, which evidenced by Fourier Transform Infrared (FTIR). Additionally, the biodegradation rate of JFSS/PVA was evaluated through soil burial test.

1. Introduction
Synthetic polymer materials are necessities, especially in the packaging industry, but they have caused major problems to the environment after being disposed as solid wastes. This is because the degradation of these materials can take up to a thousand years. Therefore, a high inclination towards the effort in developing a substitute biodegradable polymer materials have been increased interest by the researchers to overcome this problem. Having said that, numerous attempts have been made to develop biodegradable materials by adding natural polymers such as starch to synthetic polymers like poly (vinyl alcohol) (PVA) because it is well known as a synthetic biodegradable polymer and possesses excellent mechanical and physical properties [1]-[2].

Among natural polymer resources, starch is the most appealing candidates because of its low cost, pervasive availability and renewable [3]. Little information existed on modification of jackfruit (Artocarpus heterophyllus) seed starch by blending with PVA. It has been investigated that, the seeds mainly contain protein and starch, due to that, these seeds are considered as a promising source of starch [4]. Furthermore, PVA is petroleum-based polymer obtained from hydrolysis of polyvinyl acetate that is non-toxic, flexible, soluble in water, chemical resistance, good film forming and barrier properties towards oxygen [2]. Ample interest lies in blending starch with PVA because starch have demonstrated excellent compatibility in the blend [1].

This study aims to prepare biodegradable blends based on starch and PVA by solution casting method and to investigate the physical properties, tensile strength (TS) and elongation at fracture
Biodegradability studies through soil burial test were performed to investigate the effect of starch incorporation in the film’s degradation.

2. Experimental procedures
The starch/PVA blend was prepared using solution casting method according to Ismail and Zaaba [5] with slight modification. The experimental procedures were carried out on six samples coded as SP0, SP2, SP4, SP6, SP8 and SP10 which containing starch from 0 to 10 wt. %, as shown in figure 1. PVA content was fixed at 3 wt. % and the amount of starch was varied from 0 to 10 wt. % with total mass of the system is 100 g.

A mixture of 3 wt. % PVA and distilled water were heated to 95°C. JFSS was then added to the mixture and stirred continuously to 70°C to obtain good homogeneity for the mixture. Then, the film solution was casted on an acrylic plate and dried at 35°C for 24 hours. The film was peeled off from the molds and stored in the desiccator at 23°C and 30% relative humidity until further testing.

| Sample | Compositions of blends (weight percentage, wt. %) |
|--------|-------------------------------------------------|
|        | Starch  | PVA    | Distilled water |
| SP0    | -       | 3      | 97              |
| SP2    | 2       | 3      | 95              |
| SP4    | 4       | 3      | 93              |
| SP6    | 6       | 3      | 91              |
| SP8    | 8       | 3      | 89              |
| SP10   | 10      | 3      | 87              |

The mechanical properties (tensile strength and percent elongation at break) of the JFSS/PVA blend films were verified in accordance to ASTM standard D882-02 by using Universal Tensile Tester (Shimadzu; Material Testing System) with 20 mm/min of speed and 5 kN of load. The samples of cast films were cut into strips with dimension of 70 mm × 10 mm based on standard ISO–527. Next, the morphology of JFSS/PVA blend films was revealed using Field Emission Scanning Electron Microscope (FESEM; JEOL JSM-6700F). The samples were coated with a layer of gold using a coating machine (Auto Fine Coater; JEOL JFC-1600) and observed under FESEM at a voltage of 10 kV. Furthermore, the infrared (IR) spectra of the film was recorded on FTIR spectrometer (Perkin Elmer; Spectrum 100). The samples were measured at a resolution of 4 cm⁻¹ between 4000 cm⁻¹ to 600 cm⁻¹ in the average of 32 scans per sample.

3. Results and Discussion
The mechanical properties in figure 1 shows the variation of the tensile strength (TS) and percent elongation at break (E%) with different blend ratios of JFSS/PVA blend films. It is expected that pure PVA film (SP0) shows the highest TS at 19.45 MPa and E% at 57.44%, while addition of starch considerably decreased both TS and E%. The reduction of these mechanical properties probably due to the amorphous nature of starch and poor filler-matrix interaction when starch is in excess [1].

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Weight\ loss\ (%) = \frac{w_i - w_d}{w_i} \times 100
\]
increased initially for the blend films containing 4 wt. % starch. This is because higher starch amount promotes filler-filler interactions to occur rather than filler-matrix interactions [1]. After the initial increase of TS again start decreasing with increasing starch content more than 4 wt. %. Thus, considering the starch/PVA blend based on the aim of study, when JFSS was introduced, the SP4 sample gave the highest result of TS which about 10.45 MPa and optimum E% at 2.28% compared to other JFSS/PVA films. The trends obtained are in agreement with the study of rice starch/PVA blend film [6].

![Figure 1](image1.png)

**Figure 1.** Effect of increasing starch content on the mechanical properties of JFSS/PVA blend films

The morphological structure of JFSS/PVA blend films (magnification 100×) was presented in figure 2. Figure 2 (a) revealed the unfilled PVA film has smooth and transparent surfaces. When 2 wt. % of JFSS is added to PVA, the starch granules are seemed to be wetted by PVA continuous phases as shown in figure 2 (b). However, it can be postulated that due to insufficient amount of starch, it results in poor interaction between starch and PVA. Meanwhile, addition of 4 wt. % JFSS demonstrated the optimum interactions between starch and PVA compared to other blend ratio as shown in figure 2 (c). It can be observed that the starch granules are well dispersed, less agglomerated and no formation of voids took place. These interactions are responsible for high TS of the film. However, further addition of starch content more than 4 wt. % resulted in the formation of agglomerates and voids as shown in figure 2 (d) – 2 (f). The formation of agglomerate occurred because starch and PVA are not soluble when mixed together due to excess of starch in the blend [7].

![Figure 2](image2.png)

**Figure 2.** Surface morphology of (a) 0 wt. %, (b) 2 wt. %, (c) 4 wt. %, (d) 6 wt. %, (e) 8 wt. % and (f) 10 wt. % starch content in PVA blend film
The FTIR spectra of PVA and starch/PVA blend films are shown in figure 3. The wave numbers related to vibration peaks of the functional groups in PVA for each sample are depicted in table 2. All spectra exhibit the characteristic absorption bands of pristine PVA which are 3274, 2919, 1732, 1374-1243 and 1087 cm\(^{-1}\). The vibrational peaks are assigned to O–H stretching, C–H stretching, C=O stretching, C-H wagging and C-O stretching in C-O-C group of PVA, respectively. Consequently, starch/PVA blends also exhibit similar characteristic peaks in the FTIR spectra, indicating the success of blending of PVA with starch. The stretching vibration of the hydrogen bonding -OH group of PVA and starch/PVA blends was shifted from 3269 to 3274 cm\(^{-1}\). These indicate the -OH group of starches was involved in the hydrogen bond formation. The characteristic peaks at 1732 and 1716 cm\(^{-1}\) in PVA are associated to the residual acetate groups due to the manufacture of PVA from hydrolysis of polyvinyl acetate [2].

![FTIR spectra for (a) JFSS powder, (b) PVA (SP0) and (c) 4 wt% starch in PVA (SP4) films](image)

**Figure 3.** FTIR spectra for (a) JFSS powder, (b) PVA (SP0) and (c) 4 wt% starch in PVA (SP4) films

| Peak assignment | Samples' wave number (cm\(^{-1}\)) |
|-----------------|-----------------------------------|
|                 | JFSS      | SP0      | SP4      |
| C-O stretching in C-O-C | 1000     | 1088     | 1020     |
| C-H wagging     | 1367-1234 | 1374-1243| 1373-1246|
| C=O stretching characteristics carbonyl vibration in the residual acetate in PVA | "       | 1732     | 1716     |
| C-H stretching  | 2925     | 2919     | 2921     |
| -OH stretching  | 3269     | 3274     | 3274     |

The biodegradation rate of 4 wt. % starch in PVA blend film (SP4) is determined based on the respective weight loss at regular time interval (5 days) for 20 days through soil burial method. As seen in figure 4, the buried sample was suffered of weight loss, which the weight loss increased as the burial time increased. Azahari et al. [1] reported that pure PVA has the lowest weight loss about 77.19% after being buried in soil for 56 days. Whereas, the weight loss of the SP4 was found to be 27.42% after being buried in soil for 20 days. This finding was attributed to the JFSS content in the films which is believed more biodegradable than pure PVA. The high weight loss ascribed to starch/PVA blends are due to both starch and PVA content. Starch is inherently biodegradable and PVA is soluble in water. At the same time, weight loss also indicates the initial degradation process...
due to the metabolic activity of microorganisms in which they consume the constituents of the degraded material as source of nutrients [2]. In fact, the white spots observed on the surface of the film samples (figure 5) was believed due to microorganisms’ attacks. After 20 days of soil burial, it seems that the sample size become smaller, which is indicating the degradation and weight loss of the samples. Thus, it is noted that the weight loss of the sample might be continuous over time and the degradation rate of the starch/PVA is improved compared to the pure PVA.

![Graph showing weight loss of starch/PVA blend films after being buried in soil for 20 days](image)

**Figure 4.** Weight loss of starch/PVA blend films after being buried in soil for 20 days

![Macroscopic appearance of starch/PVA films after being buried for 20 days](image)

**Figure 5.** Macroscopic appearance of starch/PVA films after being buried for 20 days

4. Conclusion
The PVA blend containing jackfruit seed starch from 2 wt. % to 10 wt. % were prepared via solution casting method and their effect on the mechanical and physical properties were investigated. This study has shown that starch effectively disperse in the PVA blend and good compatibility. The optimum tensile strength 10.45 MPa and elongation at break 2.28% and form hydrogen bond interactions in PVA/starch. In addition, the results proposed that the addition of jackfruit seed starch to the PVA improved degradation behavior under natural weather compared to pure PVA.

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