Characterization of Jackfruit Straw-based Films: Effect of Starch and Plasticizer Contents

Muslimah Solehah Mohd Nazri¹, Intan Syafinaz Mohamed Amin Tawakkal¹*, Nozieana Khairuddin², Rosnita A Talib¹ and Siti Hajar Othman¹

¹Department of Process and Food Engineering, Faculty of Engineering, Universiti Putra Malaysia, 43400 Serdang, Selangor, Malaysia
²Department of Basic Science and Engineering, Faculty of Agriculture and Food Science, Universiti Putra Malaysia, Bintulu Sarawak Campus, 97008 Bintulu, Sarawak, Malaysia

ABSTRACT

Jackfruit straws are normally disposed as waste by food industries and vendors which may lead to serious environmental issue. In order to reduce the wastage and negative effects to the environment, jackfruit straw waste generated by jackfruit (Artocarpus heterophyllus) shows potential as bio-based film incorporated with starch. This work describes the effect of different starch and plasticizer contents on mechanical and thermal properties of jackfruit straw powder (JSP)/starch films. Film-forming solutions were prepared and cast by mixing JSP with tapioca starch at different ratios and for the plasticized films, ca. 15 - 40% of plasticizers including sorbitol and glycerol were incorporated into the JSP/starch films respectively. The tensile strength and modulus of JSP/starch films pronouncedly increased with increasing starch content, accompanied with a slight decreasing in the elongation at break. The result demonstrated that starch interacted with JSP, resulting in the formation of a new network to improve the properties of JSP films. FTIR spectrum analyses demonstrated the presence of hydrogen bonding in the JSP/starch film. The tensile strength of the plasticized JSP/starch films decreased with increasing sorbitol and glycerol content from 15% to 40%. However, the plasticizing effect of sorbitol became more significant than glycerol, particularly on the tensile properties and thermal stability. Thermal analysis by
thermogravimetric showed an increment in the decomposition temperature with the addition of plasticizers into JSP/starch films. The results suggest that films containing JSP and starch have the potential for the development of edible food packaging materials.

Keywords: Edible films, jackfruit straw, tapioca starch, tensile properties, thermogravimetric analysis

INTRODUCTION

Growing environmental awareness among consumers and plastic manufacturers has driven research in the development of biodegradable packaging materials. The synthesis of bio-based polymers from naturally derived resources, including polysaccharides, proteins and lipids is one of the strategies to minimize the usage of petroleum-based polymers (Kuorwel et al., 2013; Nor et al., 2017). The main drawbacks of these petroleum-based polymers is that the high degree of stability, which has resulted in a low degradability in the environment. Natural biopolymers are the best alternative due to their renewability, biodegradability and commercial viability in order to reduce waste-related environmental problems and depletion of petroleum-based polymers (Debeaufort et al., 1998).

Generally, the biopolymers of natural origins are more environmental friendly than the synthetic biopolymers and among them, starch particularly is a favourable biopolymer due to its low cost, widely accessible, exhibiting thermoplastic and biodegradability properties (Almasi et al., 2010 and González et al., 2016). This biopolymer is edible, odorless, tasteless, colourless, and constitutes a good barrier against oxygen transfer, suitable for food packaging materials including films and coatings (Dufresne et al., 2013; Yan et al., 2012). Despite the advantages of starch-based films, however, the properties of films have been limited due to the relatively low mechanical and barrier properties. By considering these drawbacks, the addition of additives, particularly plasticizer into the biopolymer matrix in order to improve the properties of the films is necessary (González et al., 2016). The flexibility of the films can be enhanced by reducing the polymer intra-molecular forces by incorporating plasticizers into the matrix. Food grade plasticizers such as glycerol and sorbitol are stable and compatible with hydrophilic biopolymeric packaging film, particularly starch-based film (Fama et al., 2005).

Jackfruit straw can be extracted from crops of jackfruit (Artocarpus heterophyllus L.), which is obtainable in Southeast Asian countries such as Thailand, Indonesia and Malaysia. Jackfruit waste, including skin, straw, core and seed accounts for approximately ca. 60-70% of its total weight (Subburamu, 1992) which can result in a major disposal problem and negative impact on the environment. At present, the available information on edible films from jackfruit straw is still limited. As ripe jackfruit straw consists of low amount of starch (13%) with slightly higher sugar content of 16% (e.g. reducing and non-reducing sugar) (Datt et al., 2008; Subburamu, 1992). These compounds may however provide insufficient properties to form films. Thus, the incorporation of jackfruit straw with biopolymer such as starch can be potentially turned this fruit waste to a valuable packaging material.
The compatibility between the main elements in a film system comprised a JSP and incorporated with starch could potentially contribute to the biodegradability and sustainability of the system as a whole. The aim of the present study is to explore the physical, mechanical and thermal properties of the JSP/starch films as a function of starch and plasticizer contents.

MATERIALS AND METHODS

Materials

The tapioca starch with the Kapal ABC brand was imported from Thailand and supplied by Thye Huat Chan Sdn. Bhd. Jackfruit of Mastura (J35) species was obtained from Perladangan Nangka Pahang, Temerloh, Malaysia.

Preparation of JSP

Ripe jackfruit straw was separated from the jackfruit peel (see Figure 1) (by discarding the core, seed and skin) and washed with distilled water to remove impurities. After washing, the straw was dried in an oven (Memmert, UF110, Germany) at 60°C for 48 h. The dried jackfruit straw was grounded into powder at 1500 rpm rotor speed using a grinder (Retsch, Cutting Mill SM200, Germany) and passed through a 0.25 mm mesh sieve size.

Preparation of Films

For films preparation, the casting technique was employed as described by Shapi’i and Othman (2016) with slight modifications. Calculated amounts of JSP and tapioca starch were mixed in 200 mL of distilled water in a beaker by using a 1:20 ratio (solid to liquid). The ratio of JSP to starch (total solid of 10 g) was varied from 8:2, 7:3, 6:4, 5:5, and 4:6. The film forming solution was first mixed homogeneously in the beaker with constant stirring using a magnetic stirrer for 5 min and then heated on the hot plate until gelatinized completely at 75°C with constant stirring for ca. 15 min. For the preparation of plasticized films, the plasticizer such as glycerol and sorbitol at different concentrations (15, 25 and
(40%) were added during the mixing of tapioca starch and JSP in a beaker with 200 mL of distilled water prior to heating. The gelatinized solution was cooled to room temperature before it was sonicated by using an ultrasonicator (QSonica LLC, Q500, USA) at 500 W, 20 Hz, 50% amplitude for 10 min by using probe with a diameter of 20 mm. Ultrasonic treatment of the film forming solution is crucial in order to remove insoluble granules in the starch matrix as suggested by Cheng, et al. (2010). This treatment can produce a good film with good transparency, improved moisture resistance and stronger structure. The sonicated film solution was then filtered by using filter paper and cast onto petri dish. The film thickness was measured with a hand-held digital micrometer (Mitutoyo 293-340-30, Japan) to the nearest 0.001 mm at four random locations on the film. No significant changes were observed for all thicknesses of JSP/starch films at different weight ratios, with an average thickness of 0.241 ± 0.007 mm.

**Colour Test**

The colour of JSP/starch films was measured using a Hænter Lab colourimeter (USP 1431 UltraScan Pro, USA) based on the CIE L* a* b* colour system, L*, a* and b*. L* describes the lightness ranging from black to white, a* and b* describe the chromatic coordinates ranging from –a: greenness to +a: redness and from -b*: blueness to +b*: yellowness. The films specimens were first placed on the surface of a white standard plate using value of L* = 97.39, a* = 0.03 and b* = 1.77 in order to calibrate the equipment (Gutiérrez et al., 2015). The measurement was recorded at three different positions for each of the films.

**Infrared Analyses**

The infrared spectral analyses of the starch, JSP and JSP/starch films were measured using a Shimadzu IR Prestige Fourier transform infrared (FTIR) spectrophotometer with an attenuated total reflectance (ATR) attachment. All spectra were recorded in the range of 550 - 4000 cm⁻¹ with a resolution of 4 cm⁻¹ and with 32 scans recorded at every point using Happ-Genzel apodization. A minimum of two random locations of the film was scanned per sample.

**Tensile Properties**

The tensile test was carried out based on ASTM D882 standard by using texture analyzer (Texture Analyzer Testing Machine 3365, USA) in order to determine tensile strength, Young’s modulus and elongation at break. The films were cut into rectangular shape with dimension of 100 × 15 mm. The initial gauge clamp and the crosshead speed was set at 60 mm and 20 mm/min respectively. A minimum of three specimens was tested for these purposes. The tensile properties were calculated from the force-deformation curves as described by Chang et al. (2000).
Thermal Analysis

The degradation and/or decomposition temperature of the films was analyzed by using Thermogravimetric Analyzer (TGA) (STA6000 Pyris 1TGA, Perkin Elmer, USA) and measured by using Pyris Series TGA 7 software. The samples were weighed ca. 14 -16 g and heated from 25 to 550°C with 10°C/min heating rate. The degradation temperature ($T_d$) was determined by the slope change that was obtained from the thermal profile curve. A minimum of two replications per sample was tested.

Data Analysis

The Excel software (Microsoft Inc., USA) was used to analyze the data whereby the statistical analysis of the data was carried out by one-way analysis of the variance (ANOVA).

RESULTS AND DISCUSSION

Films Colour

Figure 2 shows the effect of starch content on the colour of JSP/starch films at different weight ratios. It was observed that the amount of starch could have a significant effect on the colour of JSP/starch films. The JSP/starch films became lighter as evidenced by

![Graphs showing colour analysis of JPS/starch films at different weight ratios](image-url)

*Figure 2. Colour analysis of JPS/starch films at different weight ratios: (a) value L*, (b) value a* and (c) value b*
the increase of L* value (see Figure 2(a)), as the addition of starch content into the JSP/starch films increased. Tapioca starch is colourless and clear, hence the JPS/starch films with higher amount of starch demonstrated a higher lightness value. From Figure 2(b), no significant change of value a* was observed for JSP to starch ratio of 8:2 and 6:4, respectively. However, the value a* decreased at JSP to starch ratio of 5:5 and 4:6. The value b* as shown in Figure 2(c), slightly decreased with increasing starch content which caused films to appear less yellowish. It is important to note that the natural colour of jackfruit straw itself is yellow in colour, thus it may lead to the overall colour of the films (Sayuti et al., 2015). Moreover, the colour of the JSP/starch films was not affected by the presence of plasticizers particularly sorbitol and glycerol, regardless the concentrations (no data presented). This finding is in agreement with a study by Nur Hanani and Abdullah (2016) who investigated the effect of plasticizers such as glycerol, sorbitol and polyethylene glycol (PEG) incorporated into unripe banana films. The researchers reported that no significant changes were observed on the colour of banana films containing glycerol and sorbitol from 10 to 50% concentration.

FTIR

To confirm the interaction between the JSP and starch on the film, surface FTIR spectra of JSP, starch and JSP/starch films were obtained and are shown in Figure 3. Generally, it was found that the spectrum of JSP/starch film at JSP and starch ratio of 4:6 was similar to the spectrum of the JSP film. In the spectrum of all films, a broad band of –OH group was observed at approximately 3400 - 3300 cm\(^{-1}\) region. This peak corresponds to the stretching vibration of –OH group of starch and/or pectin as in the JSP film. Peak at 2921cm\(^{-1}\) is attributed to an asymmetrically starching vibration of –CH band (Deeyai et al., 2013). A sharp absorption peak at approximately 1631 cm\(^{-1}\) of starch film is observed which corresponds to the –OH stretching vibration of absorbed water in the starch film. The existence of this peak has been ascribed to the vibration of water molecules absorbed into the non-crystalline region of starch (Deeyai et al., 2013). As seen in Figure 3, there are major band changes associated with –OH groups. For instance, the presence of new hydrogen bonding in the JSP/starch film could be interpreted from the peak of –OH group (absorbed water) that shifts to 1587 cm\(^{-1}\) with a broader band than starch film (Liu et al., 2011). Interestingly, a new and prominent peak was observed from the JSP/starch and JSP film at 1741 cm\(^{-1}\) which indicates the stretching vibration of –C=O, attributes to the carbonyl group. According to Gnanasambandam and Proctor (2000), this band represents the ester carbonyl group of pectin at approximately 1760-1745 cm\(^{-1}\). It was found that the existence of this peak was weak for JSP film and no peak was present in the starch film. Moreover, the –COH banding of JSP/starch film was shifted to 1016 cm\(^{-1}\) with a narrow peak. Several others adsorption bands between 873-1471 cm\(^{-1}\) of tapioca starch film are
attributed to the contribution of various functional group such as –COC at 1143 cm\(^{-1}\) and 933 cm\(^{-1}\) which indicate the asymmetric stretching glycosidic bond and skeletal mode of α-glycosidic linkage. This result is in agreement with the study by Deeyai et al. (2013) who investigated the effect of unmodified and modified tapioca starch in atmospheric argon plasma by FTIR spectroscopy.

![FTIR spectra of (i) JSP/starch film, (ii) JSP film and (iii) tapioca starch film](image)

**Figure 3.** FTIR spectra of (i) JSP/starch film, (ii) JSP film and (iii) tapioca starch film

### Tensile Properties of JSP/Starch Films

Figure 4(a) shows the tensile properties of the JSP/starch films as a function of the starch content. Generally, the tensile strength of the JSP/starch films was affected by the starch content. It was observed that the tensile strength of JSP/starch films increased with the addition of starch content and the maximum occurred at the JSP and starch ratio of 4:6 with approximately 8 MPa. This result indicates that the starch improves the tensile strength of the JSP/starch blends due to a good compatibility of JSP and starch in nature. The tensile strength of the JSP film containing more than 6g of starch content (e.g. JSP and starch ratio of 3:7) was dropped drastically and difficult to handle during testing due to the brittle characteristic of the starch (no data presented). From this finding, the starch increases the strength of the film when incorporated into JSP film at the JSP and starch ratio of 4:6. The tensile strength of the neat tapioca starch film without plasticizer was approximately 5 MPa as reported by Shapi’i and Othman (2016) which lowered than JSP/starch films at the ratio of 4:6. Starch may provide extra contacts of hydrogen bonding between the polymers chains, responsible for the film-forming property. Furthermore, the increasing tensile strength values of the JSP/starch films, with the increase of starch ratios from 8:2 to 4:6, may be attributed due to a good interfacial adhesion and/or high formation of intermolecular hydrogen bonding between JSP and a hydroxyl group (–OH) of the starch. The presence of linear amylose structure in the tapioca starch may provide stronger and
high rigidity films (Maran et al., 2013; Mali et al., 2006). Neswati et al. (2015) prepared jackfruit straw edible films with the incorporation of 5% glycerol and 1% carboxymethyl cellulose (CMC) to improve the JSP film properties. The weak mechanical properties of pure JSP films may be attributed due to the low amount of starch in the jackfruit straw (12%) with higher content of sugar (16%) as reported by Subburamu et al. (1992).

From Figure 4(b), the Young’s modulus of the JSP film increases with the addition of higher starch content. It was found that the JSP and starch ratio of 8:2 had the lowest modulus value (4 MPa) while the film at a ratio of 4:6 demonstrated the highest value of Young’s modulus (259 MPa). This trend was attributed to the stiffening effect of the starch. Moreover, the formation of chain-chain associations in the starch film matrix may be present. According to Mali et al. (2006), the presence of hydroxyl and carbonyl group in the JSP (see Figure 3) can interact with the hydroxyl group of tapioca starch to form hydrogen bonds, and as a result, amylose gels and films are becoming stiffer and stronger. The elongation at break of the JSP/starch films at different weight ratios is shown in Figure 4(c). As expected, the elongation at break was slightly affected by the starch content. It was found that the elongation at break of the JSP/starch films behaved inversely to the tensile strength and modulus, decreasing from 30% to a minimum of 21% when the JSP

(a) Tensile Strength (MPa) vs. Ratio JSP : Starch
(b) Young’s Modulus (MPa) vs. Ratio JSP : Starch
(c) Elongation at Break (%) vs. Ratio JSP : Starch

*Figure 4. Tensile properties of JSP/starch films at different weight ratios: (a) tensile strength, (b) Young’s modulus and (c) percentage elongation at break*
and starch ratio was at 6:4. In general, the elongation at break of these films decreases with the addition of starch content. This indicates that the starch does not contribute to the elasticity of the films and this observation is likely attributed to the stiffness of the films as shown by the results in Figure 4(d). No significant change in the elongation at break of the JSP/starch films was observed with increased starch content.

**Tensile Properties of Plasticized JSP/Starch Films**

Figure 5 shows the tensile properties of plasticized JSP/starch films containing glycerol and sorbitol at different concentrations. In general, the presence of plasticizers influences the tensile properties of the JSP/starch films. It is interesting to note that the sorbitol-plasticized films exhibited higher tensile strength, Young’s modulus and elongation at break than the plasticized films containing glycerol. This finding is in agreement with a study by Nur Hanani and Abdullah (2016) who investigated the effect of plasticizer on starch-based films. From Figure 5(a), the plasticized films with glycerol and sorbitol at 15% concentration demonstrate lower tensile strength which are 4.3 MPa and 1.4 MPa respectively, than the control JSP/starch film at JSP and starch ratio of 4:6 (without plasticizer) (see Figure 4(a)). The presence of plasticizers that lowers the tensile strength may be due to their ability to reduce intermolecular forces between the polymer chains and increase film flexibility in the JSP/starch films. The sorbitol-plasticized JSP/starch films demonstrated higher elongation at break than plasticized JSP/starch films containing glycerol as shown in Figure 5(a). This may be attributed due to the higher molecular weight and greater molecule size of sorbitol that result less effective in disturbing starch-starch interaction and thus, forming a stronger film than glycerol-plasticized film (Wittaya, 2013).

From Figure 5(b), as expected, the addition of plasticizers has significantly reduced the Young’s modulus or stiffness of the JSP/starch films. It was found that the plasticized films containing glycerol at a concentration of 40% showed the lowest Young’s modulus value (2.6 MPa). Generally, the presence of starch provides high rigidity to a film because of its high intermolecular forces. Hence, with the addition of plasticizers into the starch-based films, the rigidity of the films was reduced (Sothornvit & Krochta, 2001). From Figure 5(c), it was observed that the sorbitol-plasticized JSP/starch films demonstrated higher elongation at break than plasticized JSP/starch films containing glycerol. This may be due to the high stretch ability of sorbitol than the glycerol. Moreover, sorbitol has low water-attracting ability that limits its ability to reduce JSP/starch -chain hydrogen bonding. No significant changes on elongation at break were observed for films at different glycerol concentrations. However, the elongation at break of sorbitol-plasticized films show an increment from concentration 0% to 25%, but then slightly decreased to 46% at a concentration of 40%. According to Abdorreza et al. (2011), the flexibility of the starch-based film could be increased by increasing the sorbitol content, but this can lead to crystallization of sorbitol.
in the film and thus, restricting the elasticity property. Based on these findings, plasticizer acts as film additive or lubricant able to increase the flexibility and elasticity of films (Mali et al., 2006).

![Graph](image)

**Figure 5.** Tensile properties of plasticized JSP/starch films containing 15, 25 and 40% glycerol (□) or sorbitol (■); (a) tensile strength, (b) Young’s modulus and (c) percentage elongation at break

**Thermal Analysis of JSP/Starch Films**

Thermal profiles of the JSP/starch films at different weight ratios are shown in Figure 6 and Table 1. In Figure 6, the thermal profile of the sorbitol-plasticized films at 15% and 40% loadings was compared with the control film (JSP and starch ratio of 4:6) and the results can also be observed from the derivative thermogravimetric (DTG) curves. The films have several degradation/decomposition step processes as seen in Figure 6. The thermal decomposition can occur in three main stages, which is in the first stage, an initial loss of weight was observed at temperatures between 100 and 130°C. This is due to a water loss or evaporation of water (Espitia et al., 2014; Gutiérrez et al., 2015). In the second stage, a decomposition step was observed at ca. 200 to 315 °C ($T_{d1}$), which is attributed to the decomposition of JSP and/or plasticizers (Espitia et al., 2014). The observed decomposition temperature, $T_{d1}$ may be due to the presence of pectin and/or simple sugar from the JSP itself.
that may be acting like a plasticizer. Next, the highest decomposition temperature occurred at the temperature range between 300 and 340°C ($T_d$) with the maximum decomposition temperature of 336°C, which the onset decomposition temperature of starch that undergoes oxidation (Gutiérrez et al., 2015).

Table 1 represents the decomposition temperature of JSP/starch films at different weight ratios. The result shows no significant change of the decomposition temperature with increasing starch content of the JSP/starch films. This may be attributed due to the complex interaction between the JSP and the starch content. However, the decomposition temperature of plasticized films was found to be higher than the control JSP/starch films at 4:6 ratio. This finding is in agreement with a study by Gutiérrez et al. (2015), which reported that the onset decomposition temperature of plasticized starch films would occur at a higher temperature than the neat starch due to the good interaction between the plasticizer and the starch matrix. Moreover, sorbitol-plasticized films demonstrated slightly higher

<table>
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<tr>
<th>Blends JSP:starch</th>
<th>Decomposition temperature of JSP/starch films $T_d$ (°C)</th>
<th>$T_{d2}$ (°C)</th>
<th>Decomposition temperature of plasticized JSP/starch films $T_{d1}$ (°C)</th>
<th>$T_{d2}$ (°C)</th>
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<tbody>
<tr>
<td>8:2</td>
<td>226</td>
<td>321</td>
<td>JSP/S_Sor15</td>
<td>233</td>
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<tr>
<td>7:3</td>
<td>225</td>
<td>320</td>
<td>JSP/S_Sor25</td>
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<tr>
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<td>323</td>
<td>JSP/S_Sor40</td>
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<tr>
<td>5:5</td>
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<td>317</td>
<td>JSP/S_Gly15</td>
<td>232</td>
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<tr>
<td>4:6</td>
<td>228</td>
<td>318</td>
<td>JSP/S_Gly25</td>
<td>238</td>
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Table 1

The decomposition temperature of JSP/starch films at different weight ratios

Figure 6. Thermogravimetric curves of JSP/starch films at 4:6 ratio including i) control film; ii) plasticized film at 15% sorbitol concentration and iii) plasticized film at 40% sorbitol concentration
thermally stable films than plasticized films with glycerol as shown in Table 1. For example, the decomposition temperature of the sorbitol-plasticized and glycerol-plasticized films at 15% loading was 336°C and 330°C respectively, with approximately 2% reduction. This may be due to the good intermolecular interaction between the plasticizer and the JSP/starch films.

CONCLUSION

The results of this study indicate that the colour of films, FTIR spectra, tensile and thermal properties of JSP are affected by the addition of tapioca starch. The presence of the starch increases the tensile strength and modulus of JSP/starch film. It was noted that an increased loading of starch in the JSP/starch films resulted in a decrease of decomposition temperature indicating a reduced thermal stability of the films. Sorbitol-plasticized films demonstrated higher tensile properties and thermally stable than the plasticized films with glycerol in the TGA decomposition temperature. This JSP/starch film has the potential to be used as an edible bio-based packaging material. The results of this study provide some fundamental data related to physical, chemical, mechanical and thermal properties which can be used as reference for future research work.

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