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Edible Film Innovation from Jackfruit Seed Starch (*Artocarpus hetrophyllus*) with the Addition of Sorbitol and Carrageenan

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ABSTRACT

Edible films are considered as the best alternative as an environmentally friendly and safe food packaging. Edible films could be made from plant-based (e.g. starch, cellulose and lignin) and animal-based (e.g. protein, casein and lipids) starting materials. This study investigated the production of edible films from widely available starch-based materials. In this research, the synthesis of the edible films from jackfruit seed starch was carried out by adding sorbitol as a plasticizer and carrageenan as a biopolymer. Carrageenan was added at various concentrations (0-60 wt.%). Furthermore, the effect of carrageenan concentration on the mechanical properties of the edible films (tensile strength, elongation, and modulus young) was investigated. The prepared edible films were applied on the bananas' skin to improve the shelf life of the fruit. The edible films with the addition of sorbitol plasticizer and carrageenan biopolymer could affect the tensile strength, elongation and elasticity (modulus young) values of the edible films. The increase in the tensile strength and elasticity of the edible films was observed with increasing carrageenan concentrations. In contrast, the elongation value of the edible films decreased with increasing carrageenan concentrations.

Keywords: plastics; edible films; starch; sorbitol; carrageenan

1. INTRODUCTION

In keeping the shelf life of a food product, a proper packaging that can protect the food from environmental contamination and keep its quality up to the hands of consumers is required (Kusumawati & Putri, 2013). Plastics are commonly used to be a food packaging, due its strong and lightweight properties. However, the use of plastics can potentially hazard the human's health. The toxic compounds from plastic packaging components could transfer to the food during storage. In addition, an excessive use of plastic can cause environmental problems due to its non-biodegradable properties. Micro organisms are commonly unable to change and specifically synthesize enzymes for the degradation of petrochemical-based polymers (Darni & Utami, 2009).

An alternative replacement with safe and environmentally friendly (biodegradable) properties is urgently required, i.e. edible films (Perez et al., 2006). In addition, edible films have been reported to be able to reduce the deterioration in the quality of the packaged food due to environmental, chemical and biochemical factors (Salamah et al., 2015). Edible films could be synthesized from various plant-based (e.g. starch, cellulose and lignin) and animal-based (e.g. protein, casein and lipids) materials (Setiawan et al., 2015). Synthesis of edible films from starches have been extensively studied, such as from breadfruit starch (Triwarsita et al., 2013), starch of mangrove plant fruit (Jacoeb et al., 2014), potato starch (Sessini et al., 2016), pea starch (Saberi et al., 2017), cassava and taro starches (Gutierrez, 2017).

Generally starch-based edible films has some weaknesses such as fragile, easily broken and less flexible (Mali et al., 2005). Therefore, additional materials are needed to improve this drawback. Starch-based edible films also have a low flexibility and high water vapor transmission rate (Santoso et al., 2011). Adding plasticizers in the film formulation could be done to improve these properties (Santoso et al., 2011). The commonly used plasticizier in the synthesis of starch-based edible films were palmitic acid, glycerol, and sorbitol (Anggarini et al., 2013). The use of sorbitol as a plasticizer could reduce the internal hydrogen bonds in intermolecular bonds and further inhibit the water evaporation from the products, lower the O_2 permeability properties (Setiani et al., 2013). Moreover, sorbitol is widely available and non-toxic.

In addition, the addition of a biopolymer (e.g. carrageenan) could increase the mechanical strength of the edible films (Darni & Utami, 2009). Carrageenan contains sulfate and non-sulfate groups, has a strong gel structure that allows to produce edible films with good characteristics (Jacoeb et al., 2014). It has been reported that the addition of jackfruit seed starch and glycerol affected the mechanical properties of the resulted edible films (Anggarini et al., 2013). Other studies showed that the addition of chitosan and glycerol as a plasticizer in the synthesis of the edible films from jackfruit seed starch reduced the thickness, moisture content, melting point and solubility of the resulted edible films (Hidayah et al., 2015). However, there have been no studies that examined the addition of carrageenan-sorbitol mixture as a plasticizer in the synthesis of the edible films from jackfruit seed starch. The synthesis of the edible films from jackfruit seed starch with the addition of various concentrations of sorbitol-carrageenan mixture as a plasticizer has been conducted in this study. The investigation of the mechanical properties of the synthesized edible films was also carried out.

2. METHODS

In this study, The "dulang" jackfruit seeds obtained from Gunungpati, Semarang, Indonesia were used as a feedstock. Sorbitol plasticizer and kappa carrageenan biopolymer were purchased from Merck and Lansida Group (Yogyakarta, Indonesia), respectively. This study consisted of several stages of experiments, i.e. the preparation of jackfruit seed starch, the synthesis of the edible films with various concentration of plasticizer, the characterization of the prepared edible films and the application of the edible films on bananas' skin.

The preparation of jackfruit seed starch was conducted as reported earlier (Hidayah et al., 2015). Jackfruit seeds were washed. The skin was then discarded. The jackfruit seeds were dried for 1-2 days under sunlight and cut into small pieces. The pieces of the jackfruit seeds were then soaked in water for ± 24 hours then drained. A 100 ml of distilled water was added into the jackfruit seeds for blending process resulting in a porridge-like mixture. The jackfruit seed porridge was filtered using clothes until the starch came out. The starch would be deposited in ± 12 hours. The starch was dried using an oven at 95°C for 2 hours and ready to be used at the next stage.

The synthesis procedure of the edible films from jackfruit seed starch was reported earlier (Ariska & Suyatno, 2015) with few modification. A 5 g of the starch was suspended in 100 mL of distilled water. Kappa carrageenan at various concentration (0-60 wt% of starch) and 1 g of sorbitol were added to the starch suspension. The mixture was then heated in a water bath at 85°C and stirred with a stirring rate of 225 rpm for 15 minutes until a homogeneous mixture was obtained. The homogeneous mixture was cooled to 50°C and stirred slowly to release bubbles. The mixture was poured into a film molding equipment, a 30 cm x 20 cm acrylic film printing, and flattened to obtain a film with a uniform thickness. The resulting film was dried in an incubator at 75°C for 10 hours.

The tensile strength (in MPa), elongation (in %) and elasticity (modulus young) of the prepared edible films were measured using a Brookfield CT03 4500 texture analyzer. The value of tensile strength was obtained from the maximum stress of film divided by the cross-sectional area of film (equation 1, where τ is tensile strength of a film (MPa), F_{max} is maximum stress of a film (N), and A is The cross-sectional area of a film (mm^2)). The cross-sectional area was obtained by multiplying the initial length of film by the initial thickness of the sample.

$$\tau = \frac{Fmax}{A} \quad (1)$$

The elongation value of an edible film is its ability to extend when it is given a tensile force. The elongation value of the prepared edible film was calculated using equation 2.

$$Elongation = \frac{strain\ when\ breaking\ up\ (mm)}{initial\ long} x100\% \quad (2)$$

Elasticity describes the stiffness or tendency of an object to change along an axis when it is given a tensile force. The elasticity of the prepared edible films was obtained from the ratio of tensile strength to elongation.

In addition to the mechanical properties, an identification of the functional groups in the prepared edible films was also carried out using a Perkin Elmer Spectrum 100 FT-IR spectrometer. This analysis was also aimed to identify the compounds and justify the successfulness of the synthesis of the edible films from jackfruit seed starch in this study.

This study also investigated the application of the synthesized edible films as a coating on bananas' skin. The bananas were separated from the tubers, then dipped in the edible film mixture at 50°C. The coated bananas were dried at room temperature and stored for 12 days at the same temperature. The change in color, elasticity and texture of the bananas' skin was observed. The bananas without an edible film coating also underwent the same treatment as a comparison.

3. RESULTS AND DISCUSSION

3.1. Mechanical Characteristics

Tensile strength is the maximum strain that a material can accept before breaking or tearing. The effect of carrageenan addition on the tensile strength of jackfruit seed starch films is presented in **Figure 1**.

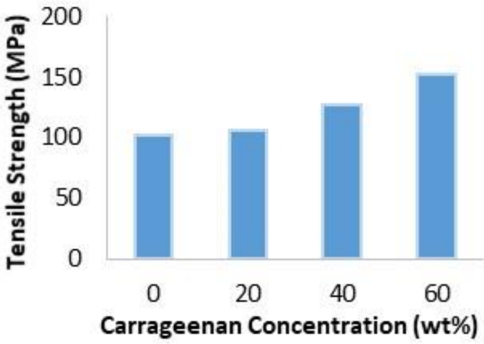

Figure 1. Effect of carrageenan addition at various concentrations (0-60 wt%) on the tensile strength of the edible films from jackfruit seed starch

Figure 1 showed an increase in the tensile strength of the jackfruit seed starch edible films. This indicated that the higher concentration of carrageenan would cause a better ability of the edible films to bind water, resulting in a gel matrix that could increase the tensile strength of the edible films (Irianto et al., 2006). Edible films that have high tensile strength will be able to well protect the packaged products from mechanical disturbance (Herliany et al., 2013). It has been shown in this study that the addition of carrageenan significantly affected the tensile strength of the edibles film produced. The minimum standard of tensile strength value of edible films based on the Japanese Industrial Standard was 3.92 MPa (Ariska & Suyatno, 2015). The tensile strength values of the edible films prepared in this study were 102, 105.39, 127.48 and 151.66 MPa for the prepared edible films with carrageenan concentration of 0, 20, 40, and 60 wt%, respectively. These value met the minimum requirement of the standard.

Elongation is the change in the maximum length of a film at the time of stretching until the film breaks. The effect of adding carrageenan at various concentrations to the elongation of jackfruit seed starch films can be seen in **Figure 2**.

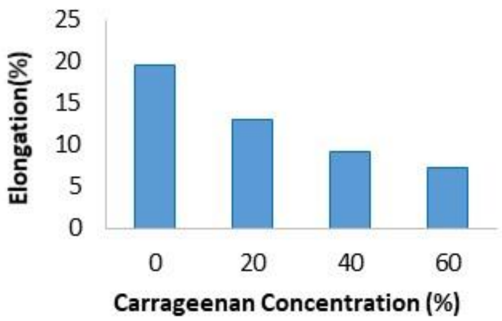

Figure 2. Effect of carrageenan addition at concentrations of 0-60 wt% on the elongation of the edible films prepared from jackfruit seed starch

The elongation of the edible films prepared from jackfruit seed starch with the addition of various concentrations of carrageenan (0-60 wt%) was observed to decrease, as is shown in **Figure 2**. The addition of carrageenan would cause a decrease in the elongation due to the formation of a film matrix that would be increasingly inelastic or easily broken (brittle) (Handito, 2011). As a result, an extension percentage of the edible films would also decrease. The value of the extension of an edible film was always inversely related to the value of its tensile strength; the higher the force required to draw the edible films, the lower the extension obtained (Cicilia, 2017). The Japanese Industrial Standard set an elongation category as poor and very good if the elongation percentage was less than 10% and more than 50%, respectively (Ariska & Suyatno, 2015). The edible films prepared in this study without and with the addition of 20 wt% carrageenan showed a fairly good elongation (19.57% and 13.09%, respectively) and met the elongation standard value.

The elasticity value (modulus young) is a measure of the stiffness of the resulting edible films. This elasticity value is obtained from a comparison between the tensile strength and the elongation of the edible films (Setiani et al., 2013). The elasticity of the prepared edible films in this study is presented in **Figure 3**.

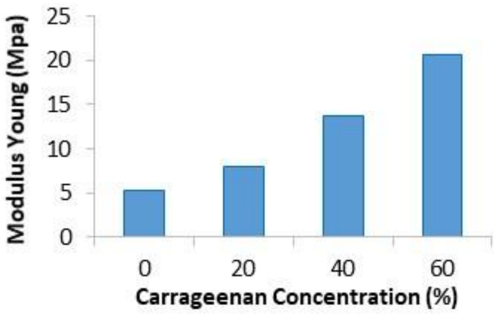

Figure 3. Effect of the addition of carrageenan at concentrations of 0-60 wt% on the elasticity of the edible films prepared from jackfruit seed starch.

An increase in the elasticity of the prepared jackfruit seed starch was observed by increasing the concentration of carrageenan as is shown in Figure 3. It was supported by studies reported earlier (Ariska & Suyatno, 2015). The value of young modulus of an edible film is directly proportional to its tensile strength and inversely proportional to its elongation. The edible films with high modulus young values could protect and coat the packaged stuff well as previously reported (Ariska & Suyatno, 2015). In this study, all edible films prepared with the addition of carrageenan (0-60 wt%) in this study had a elasticity value (modulus young) that has met the minimum Japanese Industrial Standards standard i.e. 5.21 MPa, 8, 05 MPa, 13.77 MPa and 20.63 MPa as seen in **Table 1**.

Tabel 1. Comparison of characteristics of the edible film analysis results with the Japanese ındustrial standard (JIS).

Parameter	Analysis results	JIS Standard*
Tensile Strength (MPa)	102–151.66	3.293
Elongation (%)	19.57–7.35	10–50
Modulus Young (MPa)	5.21–20.63	>0.35

*(Ariska & Suyatno, 2015)

3.2. FTIR Analysis

An FTIR analysis was performed to detect the functional groups, identify the compounds and analyzed the mixtures of samples as well as the mechanism of interaction of functional groups in mixtures. The identification of the functional groups in the edible films in this study could reveal either the physical or chemical process occurred in the sythesis (Setiani et al., 2013). The FTIR analysis involved only the sample with good mechanical (the tensile strength and elasticity) properties, i.e. the edible film with added 60 wt% carrageenan. The IR spectra of the edible films prepared from jackfruit seed starch with and without addition of carrageenan are presented in **Figure 4**. The summary of wavenumbers of the corresponding peaks is listed in **Table 2.**

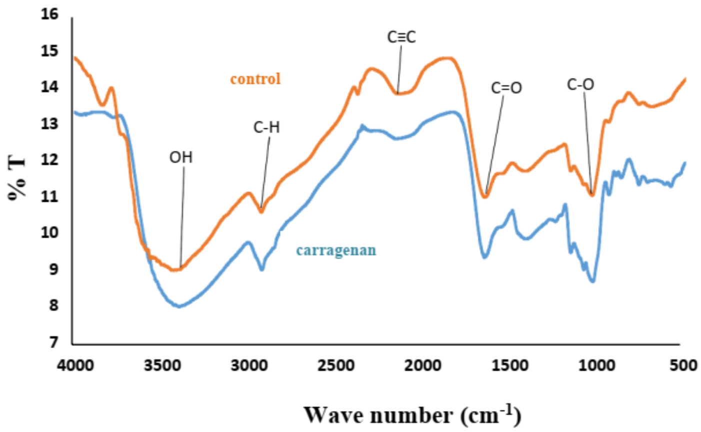

Figure 4. FTIR spectra of the edible films prepared from jackfruit seed starch without (control) and with addition of 60 wt% carrageenan.

Table 2. The summary of the wavenumbers along with the corresponding functional groups from the IR spectra of the prepared edible films.

Wavenumbers of IR spectra of the edible films (cm^{-1})		Corresponding functional groups	Possible compound
Control	Carrageenan 60 wt%		
3400,9	3422.36	OH	Phenol alcohol
2927,7	2928.68	C-H	Carboxylic acid
2155,02	2143.1	C≡C	Acetylene, alkyne
1651,05	1646.23	C=O	Carbonyl
1026,94	1030.03	C-O	Esther

Figure 4 and **Table 2** showed that the edible films with and without the addition of carrageenan contained –OH groups derived from sorbitol, carbonyl and ester functional groups. These groups were possibly derived from a combination of starch and carrageenan. The IR spectra in Figure 4 and the summary in Table 2 suggested that the prepared edible films contained O-H, C-H, C-C alkaline functional groups, C = O and C-O, without the formation of new functional groups. It indicated that the preparation process of the edible films in this study involved a physical mixing process.

3.3. Application of the Edible Films on Bananas' Skin

The prepared edible films with the addition of 60% carragenan were used as a bananas' skin coating. Coating bananas' skin using the edible film without addition of carrageenan was also performed as a comparison. The coated bananas' skin was allowed to stand for 12 days. The color, thickness and texture of the bananas' skin were observed. It was observed that the bananas coated using the edible films with addition of 60% carrageenan showed a better shelf life compared to that coated using those without carrageenan addition as seen in **Figure 5**.

The bananas coated with the edible films can last for 7 days with the insignificant change in the color of the banana (still yellow). Moreover, the texture of the banana was still good. In contrast, a sign of spoilage was observed on the uncoated banana on day 7 (Figure 5b). This was proven on the 12th day, where the banana's skin coated with the edible film was still brownish yellow, while the uncoated banana's skin had turned black with a little mold and a foul odor. Moreover, the texture and suppleness of the uncoated banana has decreased. This result was in accordance with the study previously reported (Cicilia, 2017). The reason was probably the ability of the edible film to protect the product from the possible environmental moisture and contaminant. That possible humidity and contamination would lead to the growth of microorganisms that can reduce the shelf life of the fruit products (Amaliya & Putri, 2014). The edible films would complicate the oxidation process during the storage of the fruit products because oxygen was likely difficult to penetrate through the edible film and reach the outer part of the fruit products. Therefore, the bananas coated with the edible films showed a longer shelf life than those without edible film coatings.

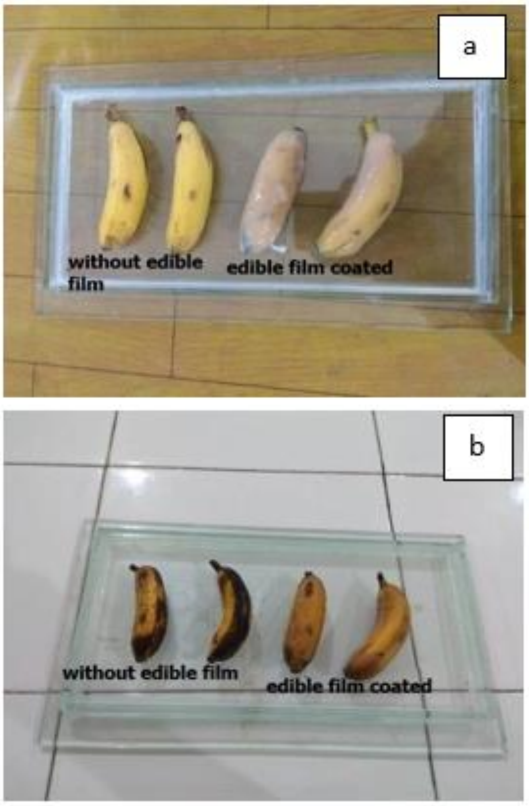

Figure 5. The appearance of the bananas with and without edible film coatings after storage on (a) day 1, (b) day 7, (c) day 12, and (d) on more than 12 days.

4. CONCLUSIONS

The addition of sorbitol and carrageenan at concentrations of 0-60 wt% in the synthesis of the edible films from jackfruit seed starch significantly influenced the tensile strength, elongation and modulus of young (elasticity) of the resulted edible films. The tensile strength and elasticity value of the prepared edible films increased with increasing carrageenan concentrations. In contrast, the elongation value decreased with increasing carrageenan concentrations. The use of the prepared edible film coatings on bananas' skin could extend the bananas' shelf life longer than those without edible film coatings.

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