Mechanical and Physical Properties of Composite Starch-Gelatin Edible Film Food Packaging

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***Abstract*—Edible films were prepared from the mixture of rice starch and fish gelatin plasticized with glycerol. This film forming solution mixture was called as composite edible films as they were prepared by combining both starch and protein material. The ratio of starch to gelatin used was varied (1:1, 1:2 and 2:1) and cast at room temperature. The edible films produced were characterized for film thickness, tensile strength (TS), percentage elongation at break (%E), water vapor permeability (WVP) and water solubility. Individual performance of either starch-based only or protein-based only were reported to have many weaknesses. This study found that addition of gelatin in starch solution has significant effect as it produce film with low tensile strength and high water vapor permeability. Composite based film showed by improving the performance in mechanical strength, water vapor permeability and other active film characteristic. Additional of glycerol also improved the film characterization performance. All tests performed on the starch-gelatin based films and focusing on producing an edible film that will have or partially have the same characteristic as synthetic plastic that were used in current market. This research also drives upon the environmentally issues of non-biodegradable waste disposal.**

Index Terms—Rice; gelatin; edible; film.

I. INTRODUCTION

In recent years, environmental fear has increased the awareness in searching the renewable and biodegradable agricultural resources to prepare packaging material. Food are obligation for us with a big contribution to stay alive. Quality and age of storage of foods will be decreased when food and environment interact. It also releasing moisture content and odor as well as triggering the growth of microbe which lead to contamination. Packaging is compulsory in order to protect and store foods from being exposed to the environment.

Edible film has a definition where a thin layer is produced on a food as a coating or packaging placed on or between the food components. Edible also means that the packaging can be consumed by human. It also can provide a good barrier to moisture, oxygen barrier and moving of solute through the packaging. Beside edible film as food packaging, it also considered as one component of the food. It should meet the properties same as synthetic packaging such as their mechanical efficiencies, physical and microbial stability as well as solubility in order to be consumed by human.

Basically, edible film is made from material with the ability to produce film. Film production includes the addition of plasticizers, antimicrobial agent, colors and flavor in the process upon the needs and requirements. The process where starch and water are regulated with heat is called starch gelatinization. The process of film formation is a condition where the gel is forming due to temperature involvement (Astuti & Erprihana, 2014). The starch granules will then swell and start thickening. As a result, the water will be absorbed slowly in an irreversible way. This gives the mixture solution a viscous and transparent texture.

Starch is the most often used by the food industry which usually used as they can provide biodegradable film to replace the plastic entirely or partially, and also can be consumed by human. Plasticizer is one of the material that widely used in the making of edible film. Plasticizers likes glycerol, sorbitol, monoglycerides, polyethylene glycol, and glucose are often used to increase flexibility and elasticity of bio-based material. This is because of alteration of their functional properties (Issue *et al.*, 2013). Composite film will overcome the disadvantage of each stand-alone film. Gelatin is used as it can improve the performance of starch-based film. Gelatin was found to be compatible with glycerol by showing the highest plasticizing effect as it can produce a flexible and ease in handling film (Vanin *et al.*, 2005).

The purpose of this study was to examine the mechanical and physical properties which included tensile strength, elongation at break, water vapor permeability and water solubility of composite starch and gelatin edible film plasticized with glycerol.

II. METHODOLOGY

A. *Materials*

Commercial fish gelatin brand Kijang was purchased from local market in Shah Alam. The halal fish gelatin was manufactured by Spicon Product Sdn. Bhd. Melaka. Rice flour was also purchased from local market in Shah Alam (Malaysia). Glycerol (98% purity), ChemAR (Germany) of analytical grade was used in this work.

B. *Preparation of composite film forming solutions*

The method to prepare the film was adopted from Al-Hassan and Norziah (2012). The film forming solution were prepared using rice flour and fish gelatin with addition of glycerol as plasticizer. Different ratios of rice flour and fish gelatin based on total weight basis (5g) including 25% (w/w) glycerol in 200 mL distilled water were prepared. Fish gelatin was dissolved in distilled water at 60˚C for 30 minutes until clear solution procured. Rice flour was dissolved in distilled water, gelatin solution and glycerol. The solution was stirred and heated to 90˚C to 95˚C for 15 minutes to promote well mixing with magnetic stirring on hot plate. The solution was approximately required 30 minutes to gelatinized. The gelatinized mixture was cooled at room temperature for 15 minutes before casting. The mixture (15g) was cast onto petri dish and dried and ventilated oven at 30˚C for 24 hours. The dry films obtained were peeled off and stored in a desiccator containing silica gel for further analysis. Control films were prepared in the same way for rice-based and gelatin-based. Each film formulation was prepared in triplicates.

C. Film Thickness

Thickness of the films was measured using digital micrometer (Mitutoyo, Japan) with an accuracy of 0.001mm. Three different positions of samples were measured and average thickness was calculated.

D. Tensile Strength and Elongation at Break

Based on method from Laohakunjit & Noomhorm (2004), the mechanical properties of films prepared were evaluated by conducting tensile strength and elongation-at-break (EAB) tests. Mechanical properties were perform using ASTM D882-10 of Tensile Properties of Thin Plastic Sheeting using universal tensile analyzer (Tinius Olsen, H50KT). Film specimen strips (3.5 cm x 2.5 cm) were cut and place in a desiccator for 48 hours prior to testing. The samples were mounted between grips with initial grip gap of 15 mm and film width of 2.5 cm. the crosshead speed is 15 mm/min with 5kN load. The results of tensile and elongation tests were expressed by MPa and percentage (%), respectively. Each test trial per film consisted of three replicate measurements.

Figure 1: Texture Analyzer

E. Water Vapor Permeability

The method to perform water vapor permeability test was adopted from Pranoto *et al*., (2005) where it was determined using a glass permeation cell by ASTM method comprising silica gel as a desiccant was covered with the film to be tested and located in a controlled desiccator. The films were sealed on glass permeation cell containing 20g of dried silica gel (%RH). After taking the initial weight of the test cup, they were placed in desiccator containing distilled water (30 ˚C, ~50±2%RH). Test cup was then weighed using an electronic balance after every 1hour interval for 8 hours. A plot of weight gained versus time was used to determine slope of linear portion that represented a steady state amount of water vapor diffusion through the film per unit of time (g/h). Water vapor permeability of films was calculated as follow:

$$WVP = \frac{L \times WVTR}{P_1 - P_a}$$

Where,

L = thickness (mm)

WVTR = slope/Area

P_1 = Partial pressure of water vapor in the air, 0.32 Kpa

P_a = Air saturated to 33% relative humidity at 25˚C, 3.173 KPa

F. Water Solubility

Water solubility method was adopted from Rachtanapun *et al.* (2009) . Film pieces were cut (2cm × 2cm) and dried in ventilated oven to obtain the initial dried weight. The sample was then immersed in the distilled water of 50 ml at 25˚C in beaker sealed with parafilm for 24 hours at room temperature (Majzoobi et al., 2015). The film was then removed and dried at 60˚C the weight was denoted as final dried weight. Solubility percentage was calculated using below formula:

$$Solubility\ (\%) = \frac{Initial\ Dried\ Matter - Final\ Dried\ Matter}{Initial\ Dried\ Matter}$$

III. RESULTS AND DISCUSSION

A. Film Thickness.

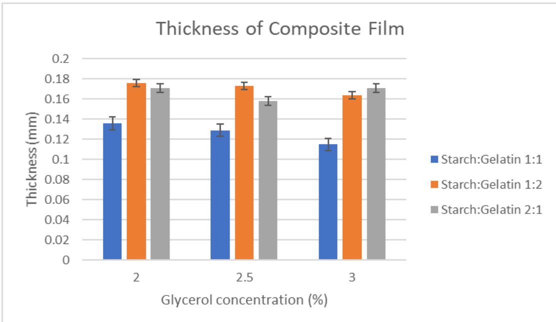

Figure 2: Thickness of composite film at different glycerol concentration

Thickness also represent in bar chart in Figure 2 where the starch to gelatin ratio of 1:1 shows decreasing and ranged between 0.1 and 0.14mm. The small different recorded by ratio 1:2 and 2:1 where the thickness are about ranged of 0.16 to 0.18mm. The thickness of composite film was measured to be thicker than the control film of starch-based and gelatin-based as showed in Figure 3 where control film showed less thickness as low as 0.08mm. This indicate that the addition of more starch or gelatinl will increase the thickness. Generally, films thickness is influence by the composition which promote the film to form and their nature of components (Ahmad *et al*., 2012). At higher concentration glycerol 3%, ratio 2:1 shows that thicker film produced compared to ratio 1:2. This is due to starch having high capacity swell of water holding and make it denser than gelatin. Thus, offered higher film-forming after the mixture solution dried (Ahmad *et al*., 2015).

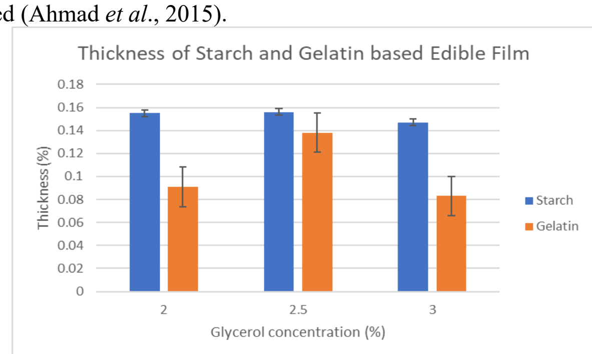

Figure 3: Thickness of starch-based and gelatin based edible film

B. Tensile Strength and Elongation at Break

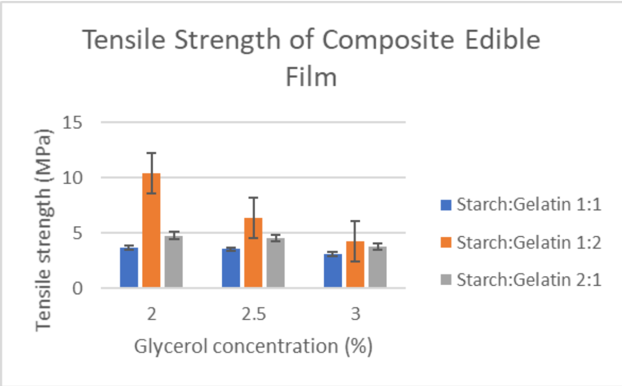

Figure 4: Tensile strength of composite edible film at varies ratio of starch to gelatin at different concentration of glycerol.

Determination of tensile strength and elongation at break of the films can give information on their applications. Different mechanical strength poses different applications of the films. Figure 4 indicates the tensile strength of composite edible film at different ratio of starch to gelatin at different glycerol concentration. It shows that increasing glycerol concentration will decrease the tensile strength at all ratio of starch to gelatin. Doubling the amount of gelatin will result in highest tensile strength at all glycerol concentration. It shows that addition of gelatin to the film solution is preferable compared to doubling the starch to the film. At ratio 1:2 the tensile strength decreases from 10.4 MPa to 4.25 MPa by increasing the glycerol concentration. Decreasing of the tensile strength was reported by Al-Hassan & Norziah (2012) as there was an interaction between hydroxyl groups with starch and protein that can reduce the interaction between starch chain.

This figure also describes that increasing glycerol influence the tensile strength of the composite film. Al-Hassan & Norziah (2012), state that higher tensile strength will make the film stronger. In this case, the higher tensile strength goes to 2% concentration of glycerol with gelatin amount doubled to starch. 2% glycerol concentration indicated that the optimum addition of glycerol to produce strong film. The addition more glycerol concentration will reduce the hydrogen bond ability to the protein chains. This was showed at the addition more at 2.5% and 3% glycerol concentration where tensile strength lower. Thus, by increasing the gelatin and glycerol will produce high tensile strength but the excessive amount of glycerol will reduce the tensile strength as the interaction between water molecules and composite edible film reduced (Azizi *et al*., 2011).

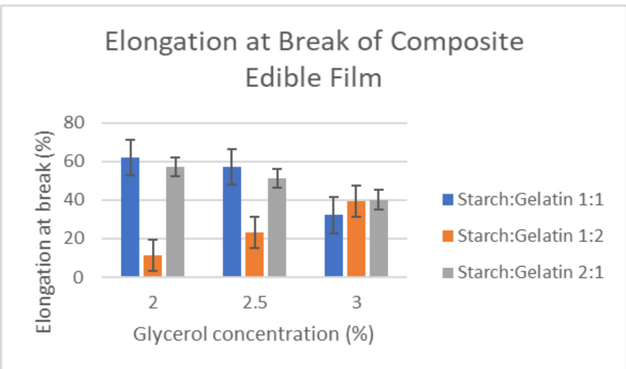

Figure 5: Elongation at break of composite edible film at varies ratio of starch to gelatin at different concentration of glycerol.

Figure 5 shows the elongation at break of composite edible film at different ratio of starch to gelatin and different glycerol concentration. At ratio starch to gelatin 1:2 shows the positive trends according to Dias *et al.* (2010) as expected. The higher the glycerol concentration the higher the elongation at break. The other two ratios of starch to gelatin show decreasing elongation at break when the glycerol concentration decrease. Glycerol will increase the effectiveness of plasticization in film forming. For elongation at break, the ratio of 1:1 show majority the highest elongation at break while the lowest elongation at break when the glycerol concentration increases to 3%. The highest elongation at break for 1:1 ratio is 61.9% at 2% of glycerol concentration.

Based on both result tensile strength and elongation at break, shows that the best glycerol concentration is 2% and ratio of 1:2 starch to gelatin reveal better performance compare to others. Higher gelatin composition will increase the effectiveness of plasticizer. Thus, will produce better tensile strength and elongation at break.

C. Water Vapor Permeability

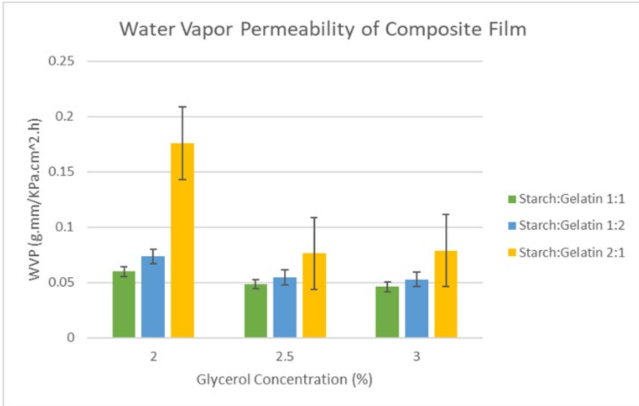

Figure 8: Water vapor permeability of composite film

Figure 8 represent the water vapor permeability of starch-gelatin based composite edible film. It shows that additional of gelatin and starch will increase the water vapor permeability. Starch to gelatin ration of 2:1 show the highest water vapor permeability compared to others in figure above. The same result was reported by Psomiadou *et al.* (1996), where the additional starch in composite film with gelatin will improve the water permeability into the films.

D. Water Solubility

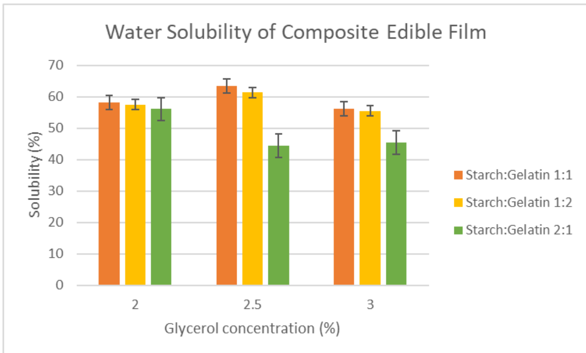

Figure 10: Solubility of composite edible film.

Figure 10 provide the data of solubility for composite edible film ad different ratio of starch to gelatin and the concentration of glycerol was also varied. The correlation shows that the higher starch ratio produces the lowest solubility of film compared to other ratio of starch to gelatin. This can be concluded that the presence of gelatin at higher amount will lead to higher solubility of film produced. While Shekarabi *et al.* (2014), mention that increasing glycerol in composite film will reduced the solubility of film. This is because the interaction of water and glycerol disturb the hydrogen bond in the film forming network. Hence, it will lead to cohesiveness reduction are the water solubility of the film itself.

IV. Conclusion

The main focus for production of composite edible film from varied ratio of rice starch and gelatin with different concentration of plasticizers to evaluate the physical and mechanical properties of the composite edible film produced. The production of composite edible film from rice starch and gelatin with different concentration of glycerol affect the mechanical properties, solubility and water vapor permeability (WVP) and microbial inhibition of the films. For mechanical properties, the composite edible films were tested by determining the tensile strength and elongation at break using tensile analyzer. From this study, there are few samples that have the potential to replace the conventional plastic packaging in market. The solubility of the composite edible film showed increased when amount of gelatin added and found to be highest at 2.5% concentration of glycerol. Higher amount of starch in composite film reduce the solubility of film towards water. As a conclusion, the addition of gelatin and plasticizers in the starch edible films brought significant impact to the mechanical properties, water solubility and water vapor permeability of the composite edible films.

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