Physical and Mechanical Properties of Composite Sago Starch-Oil Edible Film

SYLVESTER SILIN Joyce, HAMIDON Fariza
Faculty of Chemical Engineering, Universiti Teknologi Mara

Abstract—**The depleting of the sources to produce the synthetic plastics that are used widely nowadays encourages the study of the packaging of bioplastics that is made from renewable sources. One of the bioplastic is the edible film that can be produced from the starch. Despite from having wide source, edible film is one of the alternatives to produce less waste in the world. The study is done to determine the mechanical and physical of the starch-oil composite edible film that was made from sago starch with incorporation of corn oil. The edible film was prepared using five gram of sago starch mixed with glycerol (0%, 8% and 16% v/w), corn oil (0%, 2%, 4% and 6% v/w). The films were characterized for thickness, water solubility and water vapor permeability (WVP), tensile strength (TS), percent elongation at break (EB). From the analysis, all of the characteristics were affected by the additives addition in the films. The thickness, water solubility, EB and WVP increased as the amount of glycerol increased, while the TS decreased.**

***Index Terms*—Starch-oil composite edible film, sago starch, corn oil**

I. INTRODUCTION

The demand of plastics production keep on growing and this resulting in increasing usage of non-renewable resources and lead to a serious environmental pollution. Conventional plastics are basically petroleum based product. Biopolymers are produced from renewable resources and considered as an innovative solution to replace the conventional plastics that widely are used in food packaging industry (Slavutsky & Bertuzzi, 2016). The example of this biopolymer product is edible film. The edible film is a thin layer that is used to coat food, or placed between components which are relatively new and environmentally friendly food preservation technique to be compared with the plastics as the packaging materials. The source of the biodegradable materials suitable for this new idea is from natural biopolymer, such as starch.

Starch has been regarded as a promising material in edible film and coating since it has similar physical characteristics as synthetic polymers. The film produce from the starch based also transparent, odorless, tasteless, semipermeable to carbon dioxide and oxygen proof, and this properties is the same as the synthetic polymer (Vásconez *et al*., 2009). According Sessini, Arrieta, Kenny, & Peponi (2016) report, among the biomaterials present today, those derived from renewable source such as starch based products are most widespread and economical. The starch often used in food industry because it can be found abundantly and renewable. The used of starch as the material of biodegradable film is economical because it can be obtained in low cost. Sago starch is obtained from the stem of the sago palm (*Metroxylon spp.*) contains approximately 24% to 31% of amylose and the amylose content (Ahmad & Williams, 1998). Amylose component contained in starch can form a good film after drying the gelatinized amylose. According to Maizura *et al*. (2007), in gel formation, the macromolecular network is produced by inter and intramolecular network that have been formed due to the amylose and amylopectin content in the starch.

Recently, most research works on the edible materials have focused on composite or multicomponent films to explore the complementary advantages of each component as well as to minimize their disadvantages. The main target of having composite film is to enhance the film's properties to be more efficient for packaging. According to Bourtoom (2008), the composite film is the combination of either carbohydrates and lipids, proteins and lipids or synthetic and natural polymers. Galus & Kadzińska, (2015) reported that the starch-oil edible film has better moisture barrier properties compared to hydrocolloid film due to the hydrophilic lipid compound present in the structural matrix. The addition of the oil is basically used to decrease the water vapor permeability of the film. Based on work done by Garcia, Martino, & Zaritzky (2000) where the sunflower oil was used as the lipid component in the film, the water vapor permeability was decreased when the concentration of oil increased. The incorporation of starch and oil to form the composite edible film is also mainly to improve the mechanical properties of the film. As reported in the work done by Maizura *et al*. (2007), the tensile strength of the film decreased as the percentage of lemongrass oil increased.

Effective food packaging avoids the moisture transfer between the food and its surrounding or between two heterogeneous food products. Therefore,

due to the necessity, the water vapor permeability of films needs to be as low as possible. There is an optimum amount of lipid concentration in order to minimize the water vapor permeability of a film, which is related to the balancing between hydrophilic-hydrophobic ratio of a film (Garcia et al., 2000). According to Nindjin, Beyrer, & Amani (2015), the fatty acid derived from vegetable oil is consider as a safe substance with a potential to substitute the petroleum-based minerals oil and has been suggested to be used in preparation of edible film. The examples of vegetable are corn, olive, rapeseed and sunflower oil.

II. Material and Methods

A. Materials

The sago starch used was a commercialized material that purchased from the market in Dalat, Sarawak. The corn oil was obtained from local supermarket in Shah Alam, Selangor. The other materials used in this analysis were glycerol (98% purity) from ChemAR (Germany), calcium anhydrous (97% purity) and sodium chloride (99% purity) from Sigma-Aldrich (USA) obtained from Chemistry Laboratory in UiTM Shah Alam.

B. Preparation of Edible Starch-Oil Composite Film

The starch film was prepared by using previous work which was by using the casting process done by Maizura *et al.* (2007) with some modification. 5g of sago starch was mixed with 100 ml of distilled water by using stirrer at 550 rpm on the hot plate until the solution gelatinized at temperature approximately 70°C to 73°C. The glycerol (8% and 16% v/w) added once the solution gelatinized together with the corn oil was varies from 0% to 6% v/w. The mixture was allowed to mix thoroughly for 20 minutes. The 10g final solution of film was cast onto the Petri dish and dried in Froilabo oven model SN: 505137 for 24 hours at 30°C. The dried films obtained then were peeled off and store in desiccator for further analysis. Each film formulation was prepared in triplicates.

C. Thickness of Film

The film thickness is determined using Mitutoyo Digimatic Micrometer (SN: 293-230-30). Measurements were carried out at five different film locations and the average thickness value was calculated for next analysis.

D. Tensile and Elongation of Break

Tensile strength (TS) and the elongation of break (EB) test by the universal testing machine located in Polymer Laboratory, UiTM Shah Alam. Analysis of films was done according to the standard method ASTM D882-10 Films were cut into rectangular strips (35 × 25 mm). The initial gap was set at 15 mm and crosshead speed was set at 15mm/s. The data of TS and EB was measured and recorded.

E. Water Solubility

Solubility of film was tested by the method adapted from the work done by Maizura *et al.* (2007) water solubility of the films is determined by cutting the film to (2 x 3 cm) from each film. Sample was weighed to the nearest 0.0001 g and placed into beakers with 80 mL deionized water. The sample was maintained under constant agitation for 1 hour at room temperature (approximately 25ºC). The remaining pieces of films after soaking were filtered through filter paper, followed by oven drying at 60ºC to constant weight. Sample is measured in 3 replicates and the percentage of solubility is calculated as formula follows:

$$\%\ solubility = \frac{(dry\ weight)_i - (dry\ weight)_f}{(dry\ weight)_i}\ x\ 100 \qquad (1)$$

F. Water Vapor Permeability (WVP) of Film

Water vapor permeability (WVP) was determined by using the method adopted from Adjouman *et al.* (2017) where the gravimetrically method is used. In this test, film to be tested was covered the cylindrical cup containing 50 g of anhydrous calcium chloride (0% relative humidity, RH). The permeation cell was places in desiccator at 25°C that containing saturated solution of sodium chloride at the bottom of that equipment. Eight weight measurements were taken to the nearest 0.0001 g over 8 hour to record the changes in weight of the cup. A graph as a function of time has been plotted. The slope of each line was calculated by linear regression. Water vapor transmission rate (WVTR) was calculated by divided the slope (g/s) to cell surface area (m). The area of the cup was 14.52cm^2. For the calculation of WVP, it was calculated as follows:

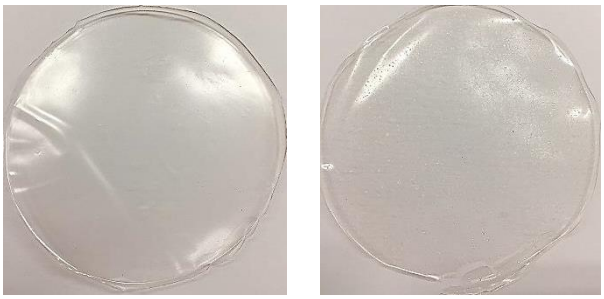
$$WVP = \frac{WVTR}{S(R_1 - R_2)} \times D \qquad (2)$$

Where, S is the saturation vapor pressure of water (Pa) at the test temperature which was 25°C. R_1 is the RH in the desiccator R_2 is RH in the permeation cell and D is the thickness of the film (m).

III. Results and Discussion

Physical properties

According to visual examination, the obtained films were transparent and they were easy to handle. However, for films incorporated with oil, they shown the droplets of oil whereas for the control film the films were clear as shown in Figure 1 below.



(i) (ii)

Figure 1: (i) Film without corn oil (ii) Film with corn oil

Thickness of an edible film is one of the important characteristic to determine the feasibility of this type of film as a food packaging material which can affect other characteristics of film such as tensile strength, elongation and water vapor permeability (Arham *et al*., 2016). Table 1 shows the thickness of the sago starch-oil composite with different amount of glycerol and corn oil incorporated.

Table 1: Thickness of the film at different amount of glycerol

Formulation of film		Film thickness (mm)
Glycerol (% w/v)	Corn oil (% w/v)	
0	0	0.0660
	2	0.0787
	4	0.0836
	6	0.0860
8	0	0.0591
	2	0.0917
	4	0.0953
	6	0.0998
16	0	0.0733
	2	0.0903
	4	0.0942
	6	0.1009

From the result obtained, for the films that contain no glycerol, the thickness is lower than the film with the glycerol. This has confirm the previous work done by Adjouman *et al*. (2017) and Jouki *et al.* (2013) where their work show the same trend which is increasing amount of glycerol will increase the thickness of the edible film. The increasing value of films thickness was due to the different concentrations of materials in each film with same amount of the film solution pour to the Petri dish before drying (Resianingrum, 2016). This causes the increased thickness in total solids of film after drying process. The work done by Arham *et al*. (2016) reported that the increase thickness of a film by the amount of glycerol is because the glycerol molecules occupied the empty space in the matrix and the interaction between the molecules and edible film. This brings the increasing between the polymers which literally cause increase in thickness.

For different amount of corn oil, the films shown an increase trend when the amount of the oil increased. Taqi *et al*. (2014) and Suput *et al.* (2016) reported that an increase oil concentration leads to an increase in the thickness of films. Valenzuela *et al.* (2013) reported in their previous work, the thickness of film increased significantly because more component were added to their edible film and they consequently conclude that thickness depend on the nature and composition of the film. Thickness of film increased especially when they incorporate with the hydrophobic molecules, which in this study is the corn oil.

Tensile Strength and Elongation of Break

Tensile strength (TS) and elongation of break (EB) are both important characteristics for packaging material. TS indicates the strength of the film meanwhile EB is the measure of film stretch ability prior to break. Table 1 is the analysis of composite film for different amount of glycerol. The TS decreased as the amount of glycerol increased. This trend was also obtained by some of previous works such as Arham *et al*. (2016) and Maizura *et al*. (2007). The reduction of TS with increasing glycerol may be due to the structural modification as the glycerol incorporated in the film. The hydrogen bond may be formed when the plasticizer interacted with the starch molecule and this weakens the interaction of starch molecule (Yunos & Rahman, 2011). Based on work done by Farahnaky, Saberi, & Majzoobi (2013), the TS significantly decreased because the matrix of the film becomes less dense, thus causing the movement of polymer chain under stress which may decrease the film resistance. However, according to Arham *et al*. (2016), the addition of glycerol may cause the decreasing of TS because of plasticizers tend to increase the moisture due to its hygroscopic nature. This causes the reduction of force between the adjacent macromolecules. The tensile strength usually decrease when the amount of lipid increase. Table 3 also shows that TS of the film decreased as the amount of corn oil increased. This result is in consistence with the results obtained by Maizura *et al*. (2007) which reported that the TS decreased significantly at all lemongrass oil concentration. This behavior may happen due to the lipid properties that act as the same as plasticizer, which increase the flexibility of the polymer chain. Based on Pranoto *et al.* (2005), the reduction of TS is reasonable in the presence of garlic oil as an additive due to the garlic oil interferes with the ionic interaction and form network.

Meanwhile for the EB, from Table 2, the percentage mostly shows increasing value. Glycerol decreases the rigidity of the network of film and increase the ability of polymer chain movement. This cause the percentage of break increased as the glycerol increases in each film. The flexibility of film is improved due to the interference of plasticizer with the polymeric chain which then facilitates their slipping properties (Farahnaky *et al*., 2013).

Table 2: Tensile strength and elongation of break of composite film

Formulation of Film		Mechanical test	
Corn oil (%)	Glycerol (%)	Tensile strength (Mpa)	Elongation of break (%)
2	0	400	4.21
	8	248	5.08
	16	137	6.93
4	0	518	5.29
	8	311	3.36
	16	103	2.79
6	0	375	1.2
	8	186	4.13
	16	104	9.27

Water Solubility

Water solubility is an essential factor to indicate the biodegradability of films when used as packaging. High water solubility means lower water resistance. Figure 2 shows the water solubility of the films. From the figure shown, the percentage of water solubility increased significantly as the amount of glycerol increased. This indicates that the plasticizer give a huge effect to the solubility of the water due to its hydrophilic property. The interaction of glycerol and water proposed the hydrogen bond. The work done by Maizura et al. (2008) also shows that the percent water solubility is higher for plasticized film. The increase trend is due to the decreasing in crystalline formation in the starch gel of the film due to the existence of glycerol. Since the formation of crystallites in film is less, the films easily swell and disintegrate, hence lower the water resistance.

There are also changes in water solubility based on the different amount of corn oil. Most of the films show a decreasing pattern as the amount of corn oil increase. This trend is reported in worked done by (Ghasemlou et al., 2013). Apparently, this trend should be expected since the oil has the hydrophobic characteristic. Hydrophobicity of the oil may cause the decrease in water solubility because this characteristic makes the film possible to retain the water. However, certain films showing different trends water solubility and this reasonable because it could be the film with the oil could be inefficient enough to resist the water.

Water Vapor Permeability

The effect of glycerol on water vapor permeability was shown in Figure 3. From the graph, in overall, the increasing value of glycerol decreased the WVP of films. Similar trend was also obtained by Maizura *et al.* (2007); in the absence of glycerol, higher WVP was observed compared to the plasticized film. Higher WVP for the film lacking in plasticizer may resulted from the brittle properties. Theoretically, the plasticizer is used to make the film more durable or flexible; therefore, if there is insufficient of plasticizer, the film would be brittle. The brittle properties enhance the microcracks occur on the film. The vapor may penetrate from the microcracks and it causes the WVP of the film without glycerol higher. By referring the work done by Maizura *et al.* (2007), where they observed that the microcracks happen on the unplasticized films meanwhile there were no microcracks on plasticized films.

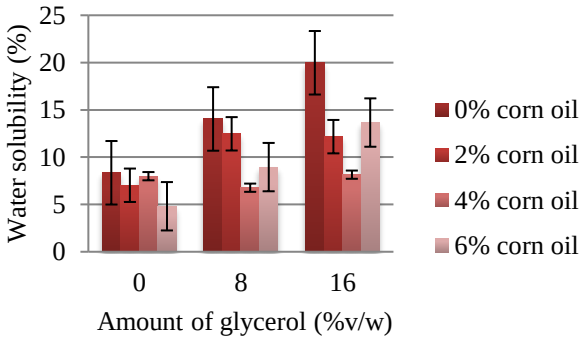


Figure 2: Water solubility of composite films

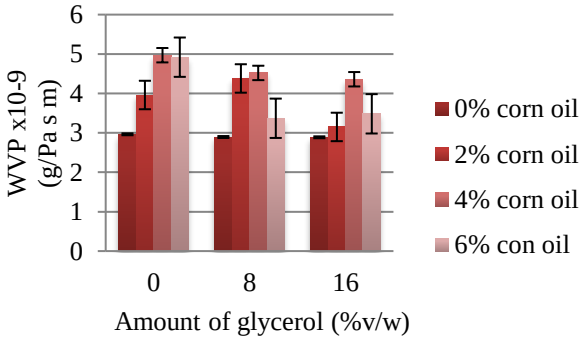


Figure 3: Water vapor permeability of composite film

The graphs also show that WVP increasing as the percentage of corn oil increased. This is due to the hydrophobicity properties as the oil was included in the film (Garcia *et al*., 2000). Slavutsky & Bertuzzi (2016)

reported the same behavior of film as the permeability of starch with lipid nanolaminated films reduced by half from film without the nanolaminated. This indicate that the lipid, the corn oil capable to repel the water vapor molecules.

IV. Conclusion

The addition of any additive such as glycerol and corn oil causes the thickness of the film increase due to the increase of the total solid of the film. The existence of hydrophobicity component which is oil caused the water solubility of the film decrease however the hydrophilicity of the glycerol could increase the water solubility. The mechanical properties of the film were affected by the presence of additives including the glycerol and corn oil. The tensile strength decreased with the addition of different concentrations of corn oil while the water vapor permeability and elongation of break increased. The glycerol was evident of the plasticizing effect and it provides the edible film with flexibility by increase the elongation of break. Water vapor permeability (WVP) of the film was also affected by the additives. The WVP increased with increasing glycerol concentration and corn oil concentration.

References

Adjouman, Y., Nindjin, C., Fa, T., Ac, D., Ng, A., & Sindic, M. (2017). Water Vapor Permeability of Edible Films Based on Improved Cassava, *8*(2). https://doi.org/10.4172/2157-7110.1000665

Ahmad, F. B., & Williams, P. A. (1998). Rheological Properties of Sago Starch. *Journal of Agricultural and Food Chemistry*, *46*(10), 4060–4065. https://doi.org/10.1021/jf980381o

Arham, R., Mulyati, M. T., Metusalach, M., & Salengke, S. (2016). Physical and mechanical properties of agar based edible film with glycerol plasticizer. *International Food Research Journal*, *23*(4), 1669–1675.

Bourtoom, T. (2008). Review Article Edible films and coatings : characteristics and properties. *International Food Research Journal*, *15*(3), 237–248.

Farahnaky, A., Saberi, B., & Majzoobi, M. (2013). Effect of glycerol on physical and mechanical properties of wheat starch edible films. *Journal of Texture Studies*, *44*(3), 176–186. https://doi.org/10.1111/jtxs.12007

Galus, S., & Kadzińska, J. (2015). Food applications of emulsion-based edible films and coatings. *Trends in Food Science & Technology*, *45*(2), 273–283. https://doi.org/10.1016/j.tifs.2015.07.011

Garcia, M. A., Martino, M. N., & Zaritzky, N. E. (2000). Lipid Addition to Improve Barrier Properties of Edible Starch-based Films and Coatings. *Journal of Food Science*, *65*(6), 941–947. https://doi.org/10.1111/j.1365-2621.2000.tb09397.x

Ghasemlou, M., Aliheidari, N., Fahmi, R., Shojaee-Aliabadi, S., Keshavarz, B., Cran, M. J., & Khaksar, R. (2013). Physical, mechanical and barrier properties of corn starch films incorporated with plant essential oils. *Carbohydrate Polymers*, *98*(1), 1–27. https://doi.org/10.1016/j.carbpol.2013.07.026

Maizura, M., Fazilah, A., Norziah, M. H., & Karim, A. A. (2007). Antibacterial activity and mechanical properties of partially hydrolyzed sago starch-alginate edible film containing lemongrass oil. *Journal of Food Science*, *72*(6). https://doi.org/10.1111/j.1750-3841.2007.00427.x

Maizura, M., Fazilah, A., Norziah, M. H., & Karim, A. A. (2008). Antibacterial activity of modified sago starch-alginate based edible film incorporated with lemongrass (Cymbopogon citratus) oil. *International Food Research Journal*, *15*(2), 233–236.

Nindjin, C., Beyrer, M., & Amani, G. N. (2015). Effects of sucrose and vegetable oil on properties of native cassava (Manihot esculenta CRANTZ) starch-based edible films. *African Journal of Food Agriculture, Nutrition and Development*, *15*(2), 9905–9921.

Paj, P., Madej, M., & Krystyjan, M. (2013). Potravinárstvo EDIBLE COATINGS AS AN ALTERNATIVE TO SYNTHETIC Potravinárstvo, *7*(March), 200–203.

Pranoto, Y., Salokhe, V. M., & Rakshit, S. K. (2005). Physical and antibacterial properties of alginate-based edible film incorporated with garlic oil. *Food Research International*, *38*(3), 267–272. https://doi.org/10.1016/j.foodres.2004.04.009

Resianingrum. (2016). Characterization of cassava starch-based edible film enriched with lemongrass oil (Cymbopogon citratus). *Nusantara Bioscience*, *8*(2), 278–282. https://doi.org/10.13057/nusbiosci/n080223

Sessini, V., Arrieta, M. P., Kenny, J. M., & Peponi, L. (2016). Processing of edible films based on nanoreinforced gelatinized starch. *Polymer Degradation and Stability*, *132*(February), 157–168. https://doi.org/10.1016/j.polymdegradstab.2016.02.026

Slavutsky, A. M., & Bertuzzi, M. A. (2016). Improvement of water barrier properties of starch films by lipid nanolamination. *Food Packaging and Shelf Life*, *7*, 41–46. https://doi.org/10.1016/j.fpsl.2016.01.004

Soltan, H. R. (2016). Extracted by Hydro-Distillationand Diethyl Ether Extraction Methods

on Four PathogenicBacteria. *Advances in Plants & Agriculture Research*, *4*(2), 2–6. https://doi.org/10.15406/apar.2016.04.00132

Uthumporn, U., Wahidah, N., & Karim, A. A. (2014). Physicochemical properties of starch from sago (Metroxylon Sagu) palm grown in mineral soil at different growth stages. *IOP Conference Series: Materials Science and Engineering*, *62*(1). https://doi.org/10.1088/1757-899X/62/1/012026

Vásconez, M. B., Flores, S. K., Campos, C. A., Alvarado, J., & Gerschenson, L. N. (2009). Antimicrobial activity and physical properties of chitosan-tapioca starch based edible films and coatings. *Food Research International*, *42*(7), 762–769. https://doi.org/10.1016/j.foodres.2009.02.026

Yunos, M. Z. B., & Rahman, W. A. W. A. (2011). Effect of Glycerol on Performance Rice Straw/Starch Based Polymer. *Journal of Apllied Sciences*, *11*(13), 2456–2459.