The Effect of Different Type of Plasticizers on Gelatin-Sago Starch Composite Edible Film For Food Packaging

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Abstract*— The objective of this study is to produce gelatin-sago starch composite edible film plasticized with glycerol and polyethylene glycol (PEG) 200, to conduct the physical and mechanical test of the glycerol and PEG 200 plasticized films, and also to determine the suitable types of plasticizers (glycerol and PEG 200) to be used in food packaging. The effectiveness of this edible film was evaluated by conducting the thickness, tensile strength and elongation at break, water vapor permeability, water solubility, FTIR, and also thermogravimetric analyses. Films were prepared by plasticizers concentration of 40%, 50%, 60% and 70% w/w of solution. Films plasticized with glycerol were thicker than films with PEG 200. This was due to the gelatin higher protein content which led to a higher viscosity and greater thickness. Glycerol plasticized films had a higher solubility compared to PEG 200 plasticized films due to the glycerol massive interaction with the gelatin-starch molecule. For the Tensile strength (TS), from the result it can be seen that the TS of GS starch film decreases with increasing plasticizers concentration from 40% to 70% and films with glycerol showed higher TS than films with PEG 200. The WVP analysis showed that the PEG 200 exhibit higher WVP than glycerol thus shows that glycerol was the best plasticizers to be used as food packaging as it had a low WVP. Meanwhile, from FTIR analysis, the absorption bands belong to the OH and CH vibrations. The thermal stability analysis showed that the glycerol TGA curves had a greater degradation rate compared to PEG 200. So the film stability in film plasticized with glycerol was higher than in PEG 200 plasticized film. But generally it indicates that the films were still stable at temperature below 100°C and still can be used for many food packaging applications. Through the conducted analysis, it can be concluded that the glycerol is the better plasticizer to be used in food packaging applications compared to PEG 200.

Keywords*— Cow *gelatin, sago starch, composite edible film, glycerol, PEG 200.

I. INTRODUCTION

Research for environment-friendly and sustainable source for packaging is gaining greater attention over the years. It is as an alternative to replace petroleum and fossil fuel based food packaging which have become the great contributor to pollutions. Composite based edible film using sago starch incorporated with gelatin is used for food packaging in this research. Films using starch have good oxygen barriers because off the tightly packed, ordered hydrogen-bonded network structure and low solubility [1]. On this basis, sago starch is a promising polymer for biofilm production due to their unique characteristics. Sago starch possesses unique characteristics but it physicochemical properties behaves much similar like common starch such as cassava and potato [2]. Besides, it is inexpensive, widely available, biodegradable, and forming odorless, colorless, nontoxic biodegradable films. It contains 27% amylose and the rest is amylopectin which is high compared to other native starches.

Gelatin is being chosen as it is the special among all of the hydrocolloids. With a melting point almost close to body temperature, it can form a thermo-reversible substance. Basically, the source of gelatin are from the skin, bones and connective tissue. It contains a unique sequence of amino acids [3]. Gelatin contain a high content of amino acids glycine, proline and hydroxyprolie. It also has a mixture of single and double unfolded chains of a hydrophilic character [4]. Starch-based films have few weaknesses. They have low resistance to water and low water vapor barrier due to its hydrophilic nature that affects its stability and mechanical properties. With the addition of biopolymers, it improves the physical and functional characteristics of starch films. Biopolymers are hydrophobic. The biopolymer use in this research are glycerol and polyethylene glycol 200 which act as the plasticizers [5].

However, glycerol and PEG 200 did increase the elongation at break but it will also increase the moisture sorption and reduced film thermal stability [6]. Therefore, polyethylene glycol (PEG) is studied too in this research as one type of plasticizer. It is non-toxic, biocompatible, non-immunogenic, non-antigenic and biodegradable plasticizer. PEG is commercially available over a wide range of molecular weights. In this study, molecular weight of PEG 200 was used. Each of the PEG molecular weight have different functions and different plasticizing effect. PEG has an excellent biocompatibility that makes it easy to be blended with other polymers. In protein starch based films, PEG will form hydrogen bonds with the protein starch chain and will reduce the intermolecular attraction, thus improve flexibility and extensibility as reported by [7].

II. METHODOLOGY

A. Preparation of Sago Starch/ Gelatin Edible Film Plasticized with Glycerol and PEG 200

The preparation of the films were performed according to the [8] method with some modifications, 5 g of sago starch was dispersed in 100 ml of distilled water producing a starch solution and was heated up with constant magnetic stirring at 75°C for 30 minutes in

a water bath until completely gelatinized. 10 g of gelatin was dissolved in 100 ml of distilled water for 30 minutes at 60°C until producing a filmogenic clear solution. The gelatin solution was added to the gelatinized sago starch solution at 60°C and stirring was continued for 30 minutes. Then, the plasticizers (glycerol and PEG 200) of 40% w/w was added to the gelatinized sago starch solution followed by constant magnetic stirring to prevent from gelatin denaturation and air bubbles for another 30 minutes. The mixture was then cooled to room temperature at 25°C and 20 ml of the solution was casted onto a petri dish and dried for 40°C for 24 hours in a ventilated oven. The rest of the films were completed by different plasticizers (glycerol and PEG 200) concentration which are 50%. 60% and 70% w/w of the solution.

B. *Film Thickness Analysis*

The film thickness was measured using the digital micrometer brand Mitutoyo with accuracy of 0.001 mm. Five different positions from each samples were taken and the average thickness of each samples was calculated [9].

C. *Tensile Strength and Elongation at Break Analysis*

Tensile strength is the mechanical property which means the maximum stress of the film before it breaks. The tests was conducted by using the tensile machine INSTRON Model 3382 by [10] method with some modification. The film samples were cut into 25 x 80 mm and was conditioned at 25°C and 55% RH for about 48 hours prior to tensile measurement. The condition film was then placed in the tensile machine grip with 40 mm initial grip separation, 2.5 kN load and crosshead speed of 500 mm min-1. The results of TS was in MPa unit and EAB unit was in percentage. Formula for calculating tensile strength was:

$$\text{Tensile strength} = F_{max}/A$$

Where F_{max} is the maximum force and A is the cross-sectional area of the film sample. The elongation at break of the film is determine by using formula:

$$\text{Elongation at break} = L_f - L_0/L_0$$

Where L_f is the final length and L_0 is the initial length of the film.

D. *Water Vapor Permeability Analysis*

According to [11] the water vapor permeability (WVP) of the films were determined using the ASTM 1989 method. The film samples was sealed on the cup containing 30 ml distilled water at 100% RH using a sealant ring and then sealed with paraffin to make sure water migration was at the exposed area only. The cups were then placed in ventilated dessicators containing silica gels at room temperature. The cells were weighed at regular time interval which is one hour for six hours when the steady state conditions were reached. Formula to calculate WVP is:

$$\text{WVP} = (\text{WVTR L}) / \Delta P$$

Where WVP is the water vapour permeability, WVTR is the water vapour transmission rate, L is the film thickness, and ΔP is the partial vapour pressure difference

E. *Water Solubility Analysis*

Water solubility analysis was conducted using [12] method with modifications. Films were cut into 2 cm x 2 cm and weighed to determine the initial weight. Then, immersed in 50 ml distilled water for 5 minutes in a beaker at different temperature which were 25° C, 40° C and 90° C. The film pieces were removed from the solution in the beaker by filtering using a filter paper to determine the undissolved dry matter of the film and were dried for 24 h at 70° C until the weight is constant. The formula to calculate solubility was:

Solubility (%) = (Initial Weight- Final Weight/Initial Weight) x100

F. *Infrared Spectrum Analysis*

The mechanism of functional group interaction that were involved on the mixture of the films were investigated using a Perkin Elmer Spectrum One FTIR Spectrophotometer with a 4 cm-1 spectra resolution. This is to determine the effects of the interaction between the gelatin and the starch and also to determine the types of functional group present in the mixture. The measurement will be repeated three times at room *temperature* [13].

G. *Thermogravimetric Analysis*

Thermal stability analysis was conducted to study the films degradation characteristics. Perkin-Elmer, TGA 7 devices by [3] method was used to determine the thermal stability of each sample. The heating rate was set to 10°C/ min in a nitrogen environment and the samples were heated at room temperature until 500 °C.

III. RESULTS AND DISCUSSION

A. *Film Thickness Analysis*

Table 1 shows the value for the thickness of the Gelatin-sago (GS) starch films. From the table shown, the thickness of the GS edible films was ranged between 0.3 mm to 0.42 mm for all films with plasticizers. Generally an increase in the plasticizer concentration led to an increase in the film thickness. This was expected as during the process of film casting, the solution became more viscous as the plasticizers concentration increase [14]. Films of GS starch plasticized with glycerol prepared were homogenous and transparent except for 60% PEG 200 (P) plasticized films. All films were found to be flexible and easily removed from the petri dishes except for the film plasticized with 60% and 70% glycerol (G). It was found to be soft, sticky and easily shrink when removed. They was then left outside for short time at lab environment and were put in the dessicator without peeling off from the plates. This may be due to the plasticizer concentration applied was more than its compatibility limit, thus causing phase separation as reported by the [15]. The G-plasticized films were thicker which was 0.41 mm than P-plasticized films which was 0.38 mm. Moreover, the gelatin consists mainly of protein, which may lead to a higher viscosity and consequently greater thickness.

Table 1: Thickness of G-plasticized films and P-plasticized films

GS film	**Glycerol, G (%w/w)**				**PEG 200, P (% w/w)**			
	40	50	60	70	40	50	60	70
Thickness	0.31	0.32	0.40	0.41	0.31	0.33	0.34	0.38

B. *Tensile Strength and Elongation at Break Analysis*

The tensile strength (TS) and the percentage elongation at break (EAB) analysis are shown in Table 2. In general, from the table it can be seen that the TS of GS starch film plasticized with both types of plasticizers decreases with increasing plasticizers concentration from 40% to 70%. When the plasticizers were incorporated into the gelatin film structure, it reduced the protein chains interaction and the proximity [16]. Films with Glycerol showed higher TS which was between 2.91 MPa to 1.46 MPa than the films with PEG 200 which was between 0.82 MPa to 0.4 MPa as the concentration increase from 40% to 70%.

Glycerol exhibit the highest tensile strength which was 2.91MPa at 40% glycerol concentration and the lowest TS which was 1.46 at

70% glycerol concentration. According to [9], glycerol exhibit more plasticization effect compared to PEG 200 which when used at the same mass content in the protein-polysaccharides based edible films. Glycerol was a smaller molecular weight structure and it was more hygroscopic compared to PEG 200. Hence it was able to insert between the protein chains and disrupts the hydrogen bonding which stabilizes the film network. Due to this, the glycerol was more effective as a plasticizers compared to PEG 200.

PEG 200 was also a good plasticizer due to the ability to reduce the intermolecular hydrogen bonding while increasing the intermolecular spacing. It contains more hydroxyl groups and interact with the water by forming the hydrogen bond [17]. From table, it can be seen that at 40% PEG 200 concentration, the TS was 0.82 MPa while at the highest concentration, the TS was at 0.40 MPa. The study reported by [18], the tensile strength and elongation at break were greatly affected by the preparation temperature and the relative humidity of conditioning. At 70% PEG 200 concentration, the TS was 0.40 MPa which was much higher than in 60% PEG 200 concentration. This was due to the films were not conditioned at a well relative humidity and temperature thus been overcome by storing the films in a dessicator with a silica gel at an accurate relative humidity and temperature.

However, the increase in the plasticizers concentration from 40% to 70% significantly increase the EAB. The EAB was the ability of the films to deform before it finally breaks. The desired flexibility of packaging films depends on their intended application. From the table 2, it clearly stated that higher EAB was the film plasticized with glycerol compared to PEG 200. At 40% glycerol concentration, the EAB is 84.83% and increase to 173.33% at 70% concentration. This increase of the EAB was due to the behaviours of the plasticizers that decrease the intermolecular bonds between the starch matrix. The reconstruction and the disruption of the starch molecular chains will reduce the rigidity and increase the film flexibility. At 60% PEG 200 concentration, it can be seen that the EAB deviate a little which it was much lower than the 50% PEG 200. This was due to the plasticizers concentration was more than its compatibility limits which had cause the phase separation [15].

PEG 200 has higher molecular weight with more hydroxyl groups than glycerol, thus it react with starch and gelatin and giving less plasticizing effect compared to glycerol. The study by [19] also reported that the gelatin actually act as a plasticizer which enhanced the film flexibility and reduce brittleness. In conclusion, the lower molecular weight of the plasticizers which was glycerol in the plasticized edible films exhibit a good plasticization effect and more suitable for food packaging application due to its higher flexibility and elasticity.

Table 2: Tensile strength (TS) and Elongation at Break (EAB) of G -plasticized films and P -plasticized films

Plasticizers Concentration (%)	Thickness (mm)	TS (MPa)	EAB (%)
40 (G)	0.303	2.91 ± 0.86	84.83
50 (G)	0.345	2.02 ± 0.86	146.67
60 (G)	0.362	1.54 ± 0.86	163.33
70 (G)	0.374	1.46 ± 0.86	173.33
40 (P)	0.306	0.82 ± 0.86	62.67
50 (P)	0.332	0.58 ± 0.86	81.50
60 (P)	0.407	0.35 ± 0.86	67.17
70 (P)	0.369	0.40 ± 0.86	106.17

*Values were given as mean ± standard deviation. G: Glycerol and P: PEG 200

C. Water Vapor Permeability Analysis

Food packaging main function was to avoid or least to decrease the moisture transfer between the food and the surrounding atmosphere or between the two components of a heterogenous product. Thus, the water vapor permeability should be kept as low as possible [11]. The film with low WVP was a good films because it can retain the moisture of the foods for a long time. In general comparison, the WVP increase significantly as the concentration increase. This was same with the study reported by [9]and [11].

From Figure 1, it can be seen that the WVP of the G -plasticized films increase which was 5.76 x 10^-6 g/mm.h.atm at 40% concentration to 6.89 x 10^-6 g/mm.h.atm at 70% concentration. This goes the same with the P-plasticized films which also increase in WVP value from 40% to 70% concentration. However, higher WVP was observed with the incorporation of PEG 200 into films compared to glycerol. This is because the free volume increases as the plasticizers was added. Thus increase the permeability. A similar behaviour was observed by [20] for gluten films added with sorbitol.

In G-plasticized films, the increase in the WVP was due to the glycerol molecules that could penetrate into the intermolecular space of macromolecules, which then facilitate the diffusion of water molecules since it is a much more smaller molecules compared to PEG 200. A large amount of water was trapped in the matrix and the swelling was promoted. So, the amount of moisture retain in the food was higher. This study was in agreement with the study report by [21]. This is in contrast with the study reported by the [9] where the order of the WVP by incorporation of plasticizers was Glycerol > PEG 200.

Gelatin was more hygroscopic than starch. Its higher affinity for water molecules led to the higher water diffusion in films and thus higher WVP. It can be concluded that the plasticizers modify the structure of the protein network and increase the WVP of edible films when both plasticizers (glycerol and PEG 200) concentrations increase. It modify the molecular organization of the protein network making films more permeable to water [22]. [18] reported that glycerol and PEG 200 is known as a plasticizers that enhance the WVP of hydrocolloid based films.

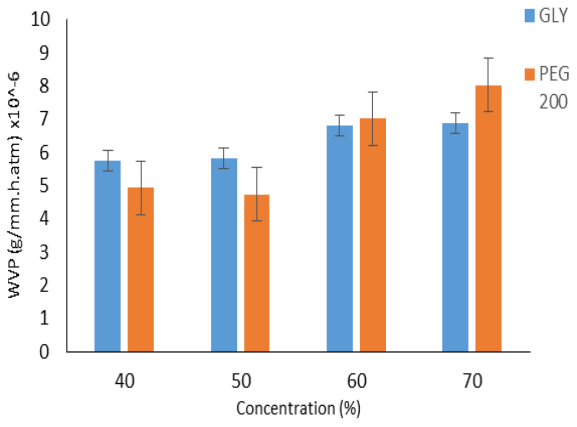

Figure 1: WVP of G-plasticized films and P-plasticized films

D. Water Solubility Analysis

The solubility tests were performed at three temperature which were 25°C, 40°C and 90°C. The G and P-plasticized films shown in Figure 2, showed significantly different values depending on the concentrations of the plasticizers. In general comparison, the solubility value of both film with increasing concentration increases with increasing temperature. As reported by [12],

increasing glycerol concentration will increase the film solubility due to the film structure changes. However, from Figure 2A shows some fluctuation of the solubility value. For example, the solubility value of 50% glycerol was higher than in 60% glycerol and same goes to figure 2B at 40°C. Besides, from Figure 2A, the solubility at 25°C also showed a drastic increase for the 70% glycerol compared to the Figure 2B which was quite constant. This was due to the film have limited tendency to interact with water molecules because of the OH groups present in its structure were involved more in the film network. This bonds will cause the stiffness and style, thus causing a lower resistance to water[23].

The Figure 2A and B also shows that the value of water solubility in film plasticized with glycerol decreases where at 70% glycerol concentration, the solubility was 89.64% at 90°C considerably in comparison with the film plasticized with PEG 200 where at 70% PEG concentration, the solubility was 96.10% at 90°C. The addition of the gelatin in both of the plasticized films had increased the water solubility and with the incorporation of plasticizers it increased the glycerol-starch interactions which interrupted the polymeric network thus increase the water permeation into the film matrix [24]. It can be concluded that the P-plasticized films exhibit higher solubility compared to G-plasticized films.

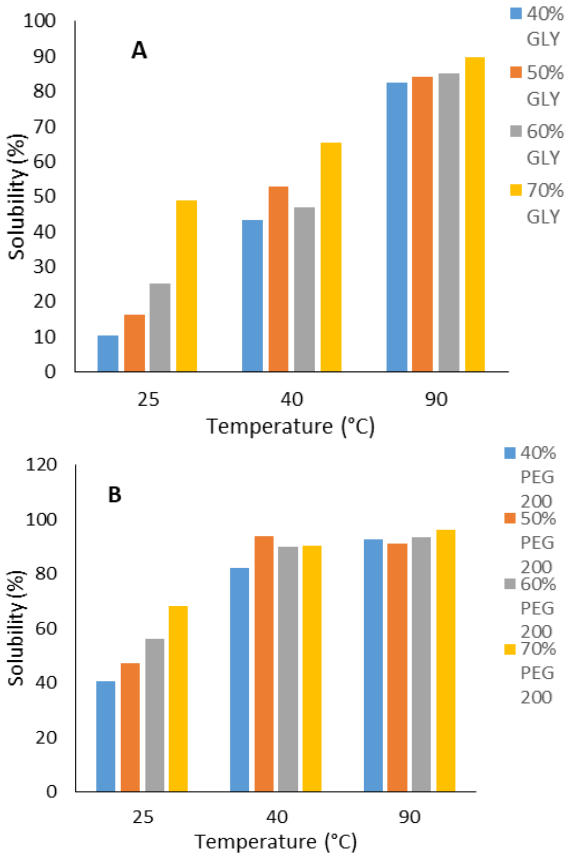

Figure 2: Solubility of G-plasticized films (A) and P-plasticized films (B)

E. Infrared Spectrum Analysis

FTIR spectroscopy was used to examine and determine the functional group present in the GS starch edible film. The FTIR spectra was shown in Figure 3A and B. The spectra for the G and P-plasticized films exhibit the same range of wavelength. The absorption bands at approximately 3290 to 3300 cm^(-1), 3100 to 2700 cm^(-1) belongs to strong and broad alcohol, OH and alkane, CH stretching vibrations, respectively. The typical spectral features for the protein were strong amide I and amide 11 bands located approximately at 1640 to 1550 cm^(-1) respectively. From the study reported by [25], the amide II absorption band was due to the coupling of the bending of the N-H bond and the stretching of the C-N bond. While in amide 1 absorption band, the band was primarily because of the stretching vibration of the C=0 bond.

Figure 3 also shows that the individual components bands in addition to the contributions of water absorptions were at 3300 cm^(-1) which was the OH stretching, 1640 which was the COH bending with abroad combination band centered around 2200 cm^(-1). The bands for starch and gelatin were identified in the spectra. It shows that the Band 1 was labelled as the saccharide bands (1064 – 883 cm^(-1)) which represent the starch region and Band 2 represent the gelatin region by the amide I and II bands.

In general, the Figure 3 shows that the increase of plasticizers concentration had increased the bands of the region and also improved the interface between the starch and gelatin molecules as reported by [25]. The FTIR suggested that for all the mixtures involved, gelatin will formed a continuous matrix in which starch inclusions were dispersed. All of the FTIR spectra showed the contributions from both starch and gelatin absorptions bands. Study showed that the PEG acted as a better compatibilizer for the starch blends than glycerol.

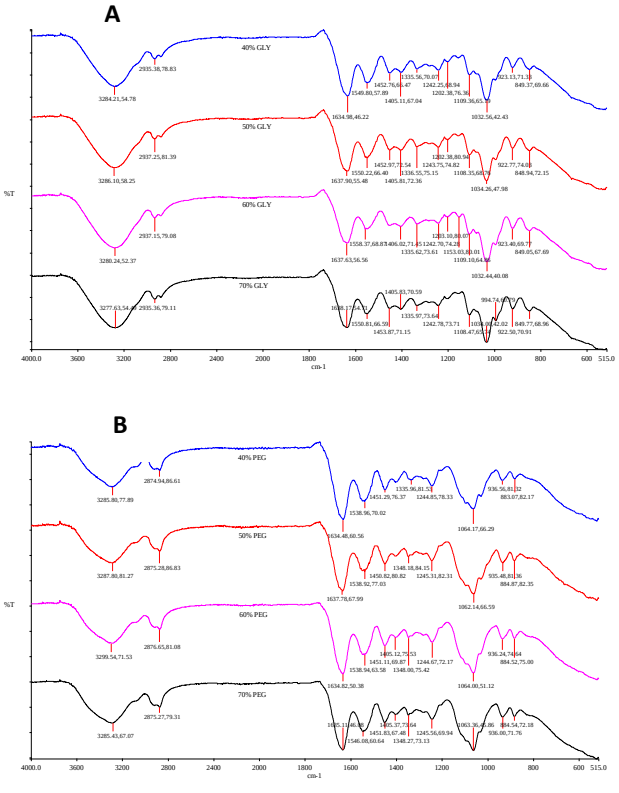

Figure 3: FTIR spectra of G-plasticized films(A) and P-plasticized films(B)

F. Thermogravimetric Analysis

This analysis techniques was used to determine the thermal stability and thermal decomposition of the plasticized films. Figure 4A and B shows the results of the TGA curves in a heating rate of 10°C/min in the temperature range from 25°C to 500°C. Based on the TGA curves shown in Figure 4, generally it can be seen that the films sample starts to degrade in a nitrogen environment at about 100°C and were fully degraded at 500°C and the thermal decomposition of the film happened in three stages (peaks).

First stage was the mass reduction which associated with the water evaporations and it occurred at temperature lower than 100°C. At this stage, the loosely bound water and low molecular weight compounds in the film were dehydrated or evaporated from the films samples [15]. From both of the curves can be seen that the film plasticized with glycerol had higher mass reduction compared to the PEG 200 plasticized films at temperature lower than 100°C. According to the [26], they reported that the mass reduction in glycerol was greater than PEG 200 due to the glycerol plasticized film exhibited hydrophilic nature with high moisture content.

For the second stage, the thermal degradation for figure 4A was in the range of 118- 327°C. This stages was associated with the evaporation of the plasticizer compounds with the water molecules. The third stage was the highest thermal degradation rate shown in both of the TGA curves occurred when heating was continued above 327°C to 500°C. This associated with the sudden mass reduction of both film plasticized with glycerol and PEG 200. During this stage, the elimination of hydrogen groups, decomposition and depolymerisation of the starch and gelatin carbon chains occurred and at this stage too, and the films were destroyed.

For figure 4A, the onset of the decomposition happened at 346°C which is not far from the one reported by [3] on glycerol plasticized films decomposition. It can be observed that the degradation rate of glycerol plasticized film increase corresponding to the increase of glycerol concentration. For example, the percentage weight left or reside left at 100°C was 37.27% for 40% glycerol, 33.44% for 50% glycerol, 34.80 for 60% glycerol and 21.74 % for 70% glycerol concentration. However the curves for 50% glycerol was higher than 60% glycerol. This may be due to the result of nucleation which sets in at higher proportion of glycerol forming crystallite that pose some resistance to thermal degradation thus reversed in favour of the curve [27].

For Figure 4B, it can be seen that the as the concentration increase, the residue of films left was much more less. This means that the higher the concentration, the higher the degradation rate which means the lower the residue left. For example is at 100°C was 89.67% for 40% glycerol, 86.00% for 50% glycerol, 89.62% for 50% glycerol and 88.29 % for 70% glycerol concentration. However for 50% PEG 200, the curve is much more lower than the other three curves. This also may be due to the result report by [27]. In comparison, the glycerol exhibit a lower thermal degradation rate than PEG 200 where at almost 500 °C, the residue left for 40% glycerol plasticized films was 75.53% while at 40% PEG 200 plasticized film is 3%. In this Figure, the second stage of thermal degradation rate occurs at temperature range of 121 - 334°C. While the third stage happens at the range of 358 - 500°C.

In other words, it can be concluded that the increase of the glycerol concentration will decrease the thermal stability of the films. This is because of the glycerol-starch-gelatin molecular interaction which weakens the strong intermolecular bonds between the starch and gelatin molecules thus lower the thermal resistance of glycerol plasticized films [28]. Comparing both of the plasticizers, the order of thermal stability of the film was: glycerol > PEG 200. But from the result shown, it indicates that the films were still stable at temperature below 100°C and still can be used for many food packaging applications. Thus, filling the gap of problem statements on the film thermal stability.

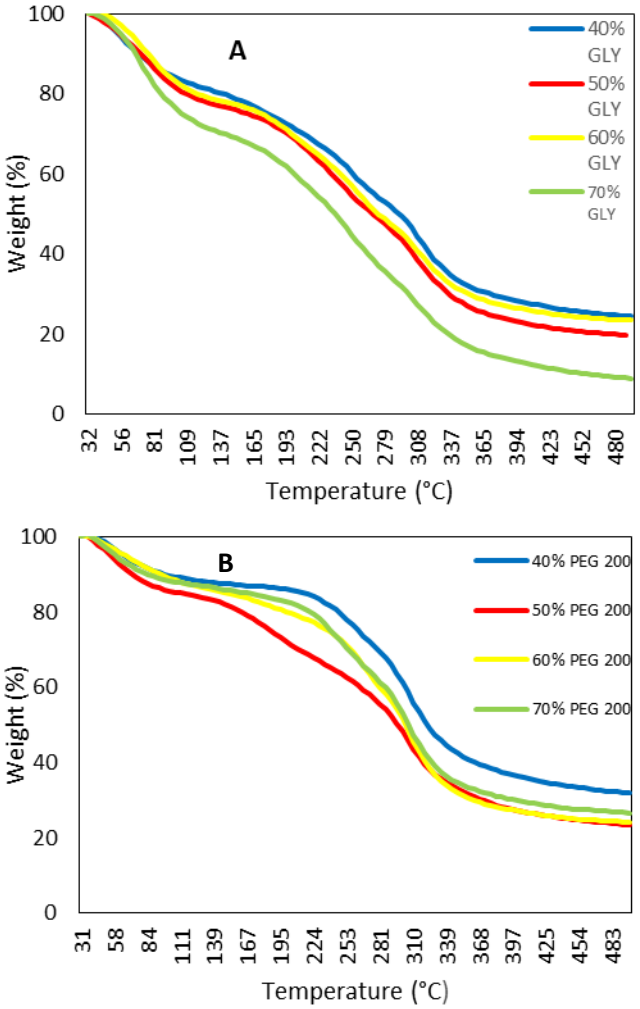

Figure 4: TGA curves of G-plasticized films (A) and P- plasticized films (B) at different concentrations (40%, 50%, 60% and 70%)

IV. Conclusion

Cow gelatin and sago starch are excellent components to make edible films because of its continuos and homogenous structure. This edible films formulated with the glycerol and PEG 200 as plasticizers were easy to handle as they were in liquid forms and not sticky after some modifications. The effect of both of the plasticizers on the physical and mechanical properties of plasticized gelatin-sago starch films depended on its concentration. Increasing the glycerol content had significantly decreased the tensile strength and thermal resistance. However, with the increase of the plasticizers concentration too had increased the solubility, thermal degradation rate, elongation at breaks, and also water vapor permeability. The films plasticized with glycerol was more suitable to be used as food packaging compared to PEG 200 plasticized films because it has a higher thermal stability. The WVP of gelatin- starch based edible films was higher for films with higher plasticizers concentration. Films with glycerol content have good flexibility and low water permeability that indicated the good edible films applications in industries especially in food and pharmaceutical.

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