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Mechanical and thermal properties of tapioca starch films plasticized with glycerol and sorbitol

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Abstract

Recently, the use of natural polymer such as starch to produce packaging film has received much attention as a substitute for conventional petroleum-based packaging film since starch is biodegradable and sustainable. However, the poor characteristic of starch such as high brittleness will limit the application of the film. This problem can be solved by adding plasticizers. In this work, the effects of different types of plasticizers (glycerol, sorbitol and 1:1 mixture of glycerol-sorbitol) on mechanical and thermal properties of tapioca starch films prepared by solvent casting method were investigated. The films were characterized in term of mechanical (tensile strength (TS), elongation at break (EAB), Young Modulus (YM) and thermal properties (thermo-gravimetric analysis (TGA)). Film plasticized with sorbitol exhibited higher TS and YM but lower EAB value which contributed to higher thermal stability than glycerol-plasticized film. This was due to the strong interactions between the polymer chains resulting from the high molecular weight of sorbitol. Hence, films became more rigid and thus promoted to a greater thermal resistance.

1. Introduction

Recently, numerous studies on the development of biodegradable film packaging especially made from biopolymers have been done. Generally, there is a huge interest in biodegradable film packaging due to the excessive use of conventional petroleum-based as food packaging material which is non-biodegradable and contributed to land disposal problems. The new study on global analysis of plastics made from petroleum reported that out of 6.3 billion metric tons of plastics have become a wastage and on top of that, only 9% has been recycled (Geyer *et al.,* 2017). In order to prevent the increasing number of plastic wastes, biopolymer-based materials have been introduced as an alternative to replace the petroleum packaging material due to its biodegradability and sustainability. Film packaging produced from biopolymers can be disposed into an organic waste accumulation for further composting, leaving behind organic by-products such as carbon dioxide (CO_2) and water (H_2O) (Othman, 2014).

The biopolymers to be used for the film formation are usually derived from renewable and abundant resources such as proteins, lipids, and polysaccharides (Nemet *et al.,* 2010). Among these biopolymers, polysaccharide mainly starch has the potential to be used for film formation due to its non-toxicity, biodegradability, renewability, availability, and low cost (Bonilla *et al.*, 2013; Li *et al*., 2015). Starch is a natural carbohydrate which serves as the main energy storage found in various plant sources such as grains (wheat and rice), fruits and vegetables (corn), as well as roots and tubers (potatoes and tapioca) (Mohammadi Nafchi *et al.,* 2013; Borges *et al.,* 2015). Tapioca starch (*Manihot esculenta* C.) was chosen to be used in this study because it can be easily found in tropical climate country such as Brazil, Thailand, Indonesia, and also in Malaysia. Films made from polysaccharides mainly starches are able to form a continuous polymer matrix even without the addition of plasticizer (Cerqueira *et al.*, 2011).

However, tapioca starch films produced without plasticizer are usually brittle and exhibit poor mechanical strength as well as thermal stability. This is due to the strong hydrophilic behavior of starch makes this material sensitive when coming into contact with water resulting in low flexibility and yield a brittle film (Avella *et al.,*

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2005). In order to overcome the film brittleness, plasticizer can be introduced as an additive in the tapioca starch film. The incorporation of plasticizer into the starch film is to enhance the flexibility of starch by reducing the intermolecular forces within polymer matrix as a result of increasing the mobility of polymer chains (Sanyang *et al.,* 2016). Plasticizers consist of hydrophilic compounds such as polyols (glycerol and sorbitol) are commonly used in starch film formation and has been found to be particularly effective to serve as plasticized hydrophilic polymer (Zhang and Han, 2006).

A number of research has been conducted regarding the effect of polyols plasticizers especially glycerol and sorbitol to the properties of biopolymer starch film. Recent studies by Lagos *et al*. (2015), investigated the mechanical properties of tapioca starch films added with glycerol and sorbitol at different relative humidity conditions. Borges *et al.* (2015) also studied the influence of different starch sources and plasticizers (glycerol and sorbitol) on the properties of biodegradable films. Although there were several studies which investigated the effect of polyols plasticizers on the tapioca starch films, the studies mostly focused on a few properties such as mechanical, physical, and barrier properties. To the best of our knowledge, this study was conducted to investigate the relationship between mechanical and thermal properties on different types of tapioca starch-plasticized films. Furthermore, there were limited studies reported on the incorporation of varying plasticizers in tapioca starch compared to other biopolymer starch-based films. In this study, the effects of different types of plasticisers (glycerol, sorbitol, and 1:1 mixture of glycerol-sorbitol) on the mechanical and thermal properties of tapioca starch films were investigated.

2. Materials and methods

2.1 Materials

Tapioca starch was purchased from LGC Scientific Sdn. Bhd. Selangor, Malaysia. Food grade glycerol and sorbitol were used as film plasticizers and were supplied from R&M Chemicals, United Kingdom with a molecular weight of 92.10 g/mol and 182.17 g/mol respectively. Distilled water served as the solvent for preparing film solutions.

2.2 Preparation of tapioca starch films with different types of plasticizers

The films were prepared by diluting 25 wt. % (based on dry basis of tapioca starch) of different types of plasticizers which were glycerol, sorbitol, and 1:1 mixture of glycerol and sorbitol (glycerol-sorbitol) in 100 mL of distilled water. After that, 3.0 g of tapioca starch was added into the plasticizer solution and then stirred using stirrer hot plate (Favorit, Model HS0707V2, Malaysia) at 80ºC for 30 mins to completely gelatinize the starch. The gelatinized starch-plasticized solution was then allowed to cool down at room temperature before sonication using ultrasonic probe (QSonica, USA) with 50% amplitude for 20 mins. Then, tapioca starch films were obtained by casting method, whereby 40 mL of the gelatinized solution was poured into 14 cm diameter of petri dish and dried in the laboratory under air conditioning system at 20ºC for 48 hrs. The sample film was then separated from the petri dish and kept in a desiccator (25ºC, RH = 53%) for further testing. The thickness of each film was measured at five different positions using Mitutoyo Digimatic Micrometer (Model MDC-1" SX, USA). The thickness of the films produced ranged from 0.007 to 0.008 mm. All the analyses were carried out at least in triplicate.

2.3 Characterization of films

2.3.1 Mechanical properties

Mechanical properties of tapioca starch-plasticized films which include tensile strength (TS), Young modulus (YM), and elongation at break (EAB) were determined using TA XT Plus Texture Analyzer (Texture Technologies, UK). Tapioca starch-plasticized sample films were cut into rectangular shape (100 x 15 mm^2) using film specimen cutter following the ASTM standard of D882 (ASTM, 2001). The samples were clamped between the grips and the experiment was run with the clamp distance of 50 mm and the cross-head speed at 0.20 mm/sec. TS is the measurement of maximum load that the sample can withstand before failure while being stretched by the texture analyzer. It was calculated by dividing the maximum load for breaking the film by its cross-sectional area. Percentage of the EAB was expressed as the change in the original length of the sample at the break and was determined by dividing the film elongation at rupture with the initial grip separation. The rigidity of the films was analyzed by the measurement of film stiffness which attributed to YM value.

2.3.2 Thermal properties

Thermal properties of tapioca starch-plasticized films were investigated *via* thermo-gravimetric analysis (TGA) using Thermo Gravimetric Analyzer (Mettler Toledo, Model TGA/DSC 1 HT, United States). At first, sample film was weighed to approximately 10.0 mg. The film was placed in aluminium pans and was heated at a constant rate of 10ºC/min from 25-400 ºC under 50 ml/min of nitrogen atmosphere. The degradation temperature (ºC) and percentage weight loss (%) were determined from two types of curve; thermo-gravimetric

analysis (TGA) and derivative thermo-gravimetric (DTG).

2.3.3 Statistical analysis

The statistical analysis of the obtained experimental results was performed by using Minitab software (Minitab Inc, USA). Analysis of Variance (ANOVA), Tukey mean comparison test (p<0.05) was performed to verify the significant differences among the mechanical values. T-test (p<0.05) was performed to verify the significant differences among thermal values.

3. Results and discussion

3.1 Mechanical analysis

Films packaging need to withstand the normal stress applied during the application, handling and storage period in order to maintain the quality of the films and food's properties. In this work, three types of plasticizers (glycerol, sorbitol, and 1:1 mixture of glycerol-sorbitol) have been used to produce tapioca starch-plasticized films. Table 1 shows the TS, EAB, and YM for different types of tapioca starch-plasticized films (control film without plasticizer, glycerol-plasticized film, sorbitol-plasticized film, and glycerol-sorbitol-plasticized film). Tensile strength is defined as the maximum tensile stress that a film can sustain during the tension grip test whereas elongation at break is the maximum change in length of the film before breaks (Bourtoom and Chinnan, 2008; Arham *et al.*, 2016). Elongation at break is usually inversely proportional to tensile strength whereby the increase in TS value will decrease the EAB.

Table 1 shows that control film exhibited the highest TS of 33.29 MPa. However, TS value decreased with the addition of plasticizers. The addition of plasticizer reduced the tensile strength of the films due to the interaction between the starch polymer and plasticizer molecules which weaken the hydrogen bonding thus, promoted the molecular mobility of the polymer matrix. Increasing molecular mobility between polymer chains resulted in the increase in film flexibility as well as film elongation which then made the film less brittle (Lin and Tung, 2010).

Table 1. Tensile strength (TS), elongation at break (EAB), and Young modulus (YM) of different types of plasticized tapioca starch films

Plasticizer	TS (MPa)	EAB (%)	YM (MPa)
Control	33.29±0.35[a]	12.54±0.20[a]	1604.07±209.11[a]
Glycerol	15.84±2.81[b]	50.21±14.09[b]	153.04±39.66[b]
Sorbitol	18.92±0.98[b]	23.21±3.54[c]	779.39±86.32[c]
Glycerol+	14.80±4.35[b]	29.70±7.27[c]	677.38±220.32[c]

Values were mean ± standard deviation. Different superscripts mean statistically different (p<0.05).

TS value for sorbitol-plasticized film (18.92 MPa) was higher than glycerol-plasticized film (15.84 MPa) due to the rigidity of sorbitol that gave strong effect to the tapioca starch film (p<0.05). According to Mali *et al.* (2005), the firmness of sorbitol plasticizer was influenced by several factors such as molecular weight and molecular size of sorbitol. They reported that the molecular weight of sorbitol was higher compared to glycerol. The high molecular weight of sorbitol plasticizer caused an increase in molecular mobility of amorphous and crystallization of the starch matrix. Therefore, films produced with the addition of sorbitol were strong and rigid than glycerol-plasticized films due to the molecular size of sorbitol which was higher than glycerol. As a result, the incorporation of sorbitol in the film was lower than glycerol plasticizer. This condition makes the film less flexible and more rigid compared to glycerol. Other than that, bigger size of sorbitol molecule affected molecular ring conformation of sorbitol molecules which strictly hindered an insertion between starch chains. As a result, sorbitol plasticizer was less effective in disturbing the interruption between starch molecules which resulted in a strong film (Wittaya, 2013).

Apart from that, TS of films incorporated with glycerol plasticizer exhibited the lowest value of TS due to the hygroscopic character of glycerol which led to an additional water to bind with starch matrix thus reducing the strength of the films (Mali *et al.*, 2005). Meanwhile, TS value for glycerol-sorbitol-plasticized film was in between the glycerol and sorbitol-plasticized films which was 14.80 MPa because the combination of these plasticizers resulted to balancing the low TS of glycerol plasticized film and high TS of sorbitol plasticized film (Sanyang *et al.*, 2016). They also reported that a combination of glycerol and sorbitol plasticizers improved the low tensile strength of glycerol-plasticized film and compromised the high tensile strength of sorbitol-plasticized film.

Table 1 also shows that control film exhibited the lowest EAB value of 12.54% followed by sorbitol 23.21% and then glycerol-plasticized film with EAB value of 50.21%. These results were consistent with the study by Borges *et al.* (2015) who found that films from potato and tapioca starch exhibited lower TS and higher EAB values when prepared with glycerol compared to those produced with sorbitol. This finding shows that glycerol-plasticized film has a greater plasticizing effect than sorbitol whereby glycerol can soften the film and make it more elastic than sorbitol due to its smaller size, which allows it to exert a greater influence on the EAB than the molecule of sorbitol (Heydari *et al.,* 2014). Besides, Dias *et al.* (2010) discovered that the highly
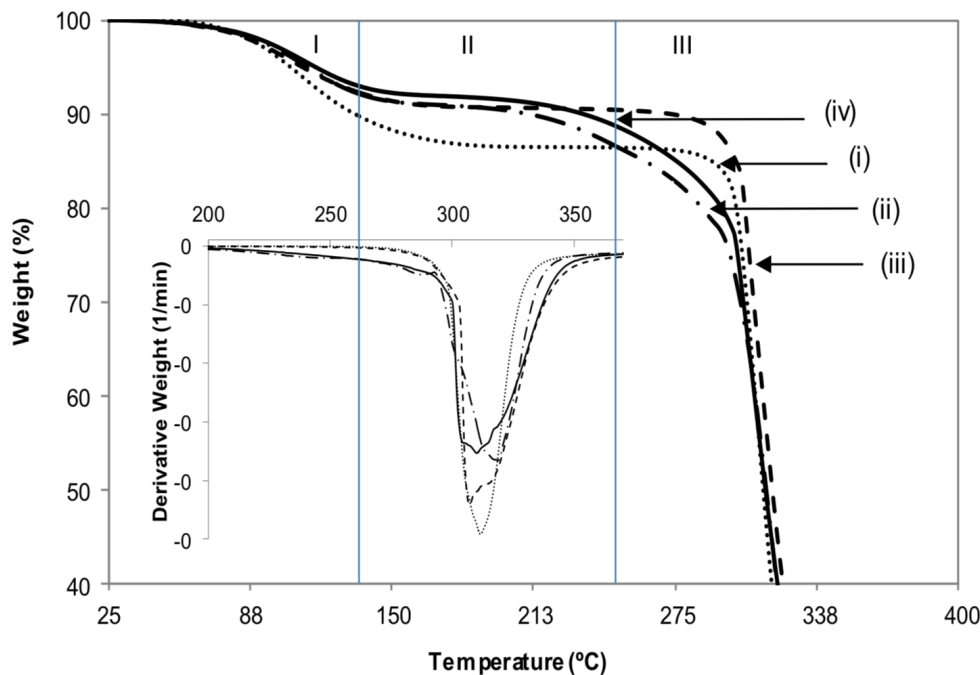
Figure 1. Thermo-gravimetric curves for different types of tapioca starch films including i) control film; ii) glycerol-plasticized film; iii) sorbitol-plasticized film; and iv) glycerol-sorbitol-plasticized film.

hygroscopic nature of glycerol-plasticized films was also plasticized by water since water is one of the plasticizing agents in starch-based materials, which explains its low TS and high EAB values. In contrary, the addition of sorbitol plasticizer reduced EAB of the film due to the molecular structure of sorbitol that was similar to glucose units, which promotes the interaction within polymeric starch chains thus, resulting to the increase in strength and stiffness of the film with low flexibility (Ooi *et al., 2012*). From Table 1, EAB value for glycerol-sorbitol-plasticized film showing a similar trend with TS value which was in between the glycerol and sorbitol plasticized films. Generally, as the film structure softens, TS decreases and EAB increases. A higher EAB value indicates that the film is more flexible when subjected to tension or mechanical stress. This implies that the film might resist mechanical damage when subjected to a rough handling during application on food processing especially for easily damaged food products (Aisyah *et al., 2018*).

YM is the measure of film stiffness, which is the extent to which films resist deformation when the response to an applied force. The higher the YM, the higher the stiffness and rigidity of the materials. Based on Table 1, YM for control tapioca starch film was the highest amongst the other films which were 1604.07 MPa. Film stiffness can be related to its TS by which the stiffness of control tapioca starch was the highest and directly proportional to its strength. When plasticizers were added, films rigidity were reduced as the YM values showed a decrement to 779.39, 677.38, and 153.04 MPa with the addition of sorbitol, mix of glycerol-sorbitol and glycerol, respectively. Prakash Maran *et al.*

(2013) have also reported that the glycerol-plasticized film was less stiff than sorbitol. This was due to its intensively hygroscopic characteristic as compared with the sorbitol, which acted as a lubricant to improve the movements between polymer chains. As a result, there was a significant increase in EAB and reduction in YM values for glycerol-plasticized film. YM value for the glycerol-sorbitol plasticizer was in the middle between glycerol and sorbitol similar to the trend of TS and EAB values as reported by Sanyang *et al.* (2016) whereby the mixture of glycerol and sorbitol plasticizers gave a balance effect to the flexibility and rigidity of the films.

3.2 Thermal analysis

In this study, thermo-gravimetric analysis (TGA) was used to determine the thermal decomposition or degradation and stability of different types of tapioca starch-plasticized films. The results of thermal degradation for different types of tapioca starch-plasticized films were presented in plot curves of weight loss (%) and derivative weight loss of the films as a function of temperature (°C) as shown in Figure 1. From the TGA curves, it can be seen that the weight loss of the films decreased with the increase in temperature which shows that the degradation of the films occurred when heated.

Control film clearly demonstrated two-steps weight loss at particular degradation temperature range of 73-151°C and 280-370°C with respect to degradation of weakly bound water and decomposition of polymeric starch material respectively (Pelissari *et al.,* 2009). However, glycerol-plasticized film shows three-step weight loss which varied in the temperature range of 68-

145ºC, 205-260ºC, and 290-380ºC, ascribed to degradation of weakly bound water, glycerol plasticizer degradation, and polymeric starch decomposition (Cyras *et al.*, 2008; Sanyang *et al.,* 2016).

According to Sanyang *et al.* (2016), the degradation temperature of unplasticized and plasticized sugar palm starch (SPS) films for glycerol occurred between 125 to 290ºC indicated by the second step. They reported that the three-step thermal degradation system also demonstrated by the film with glycerol-sorbitol plasticizer by which the degradation of glycerol takes place at the temperature range between 220-275ºC might be due to the contribution of glycerol content in the film. Zhong and Li (2014) also found that the degradation temperature of glycerol-rich phase of kudzu starch-based edible films occurred at the second step of temperature ranges from 150 and 280°C. The trend of these results can also be observed from the derivative weight loss curve which shows a slight slanted slope starting from 200°C where the degradation of glycerol happened at this step. The thermal degradation rate of polymeric material was at its highest when further heated to a temperature more than 280ºC whereby the drastic weight reduction of all films can be seen in Table 2.

Table 2. Maximum degradation temperature, Tmax (ºC) and weight loss (%) for different types of plasticized tapioca starch films at third step of thermal decomposition. P>0.05 according to statistical analysis.

Plasticizer	Degradation temp (ºC)	Weight loss (%)
Control	299	17.13
Glycerol	296	23.05
Sorbitol	304	16.13
Glycerol+Sorbitol	300	22.21

Similar trends were also discovered by various studies from Cyras *et al.* (2008); Nascimento *et al.* (2012); Suppakul *et al.* (2013); Dang and Yoksan (2015); and Sanyang *et al.* (2016). They discovered that the mass loss at the initial step can be associated by the loss of weakly bound water and low molecular weight compounds of the film, whereby the degradation of the molecular chain of plasticizer occurred at the second step and the thermal degradation for the last step was subjected to the elimination of hydrogen groups, decomposition, and depolymerization of the starch carbon chains.

It can be observed in Figure 1 that there was no significant difference (p>0.05) on the weight loss of maximum degradation peaks among the films. However, glycerol-plasticized film contributed to the lowest thermal degradation rate among all at the last step of polymeric degradation temperature between 280 and

370ºC. From Table 2, the maximum weight losses at the polymeric degradation temperature were 17.13%, 23.05%, 16.63%, and 22.21% for control, glycerol, sorbitol, and glycerol-sorbitol-plasticized films, respectively. In other words, the presence of glycerol in tapioca starch films reduced the thermal resistance due to the molecular interaction between glycerol and starch compound which weakened the strong intermolecular bonds between starch molecules and lowered the thermal resistance of glycerol-plasticized film (Sanyang *et al.,* 2016). Based on Figure 1, sorbitol-plasticized film had the highest thermal stability than other films at respective temperature. These results resembled the mechanical analysis whereby sorbitol-plasticized film also showed the highest tensile strength among others due to the sorbitol content which contributed to the optimum thermal stability.

4. Conclusion

It was found that control tapioca starch film exhibited low EAB but high TS values. Nevertheless, the addition of plasticizers in the tapioca starch films alters the mechanical properties of the film. Incorporation of high molecular weight plasticizer particularly sorbitol slightly increased the TS of the films but reduced the elongation and flexibility as compared to glycerol. Similarly, sorbitol-plasticized film also exhibited the highest thermal resistance than glycerol because the presence of high molecular weight of sorbitol in between starch polymer chain produced a stronger film due to the decrease in molecular mobility of the polymer chains thus improved the thermal stability.

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