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Effect of Plasticizers on Physicochemical and Mechanical Properties of Chitosan-Gelatin Films

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Abstract. Composite chitosan-gelatin films were produced to investigate the effect of plasticizer and composition of chitosan and gelatin on physicochemical and mechanical properties of the films. The films were prepared according to ratio of chitosan: gelatin of 1:1, 1:2 and 2:1. For each film, glycerol, sorbitol and sucrose were added as plasticizer. The film forming solution was poured on a glass plate and dried for 12 hours in an oven at 60°C. The highest tensile strength was 4.04 MPa for films of ratio 2:1 plasticized with glycerol compared to sorbitol and sucrose which were 3.94 MPa and 3.84 MPa, respectively. However, films plasticized with sorbitol at ratio of 1:2 had the highest percent elongation which was 68.20%, followed by glycerol and sucrose which were 26.51% and 24.08%, respectively.

1. Introduction

Synthetic plastics are non-biodegradable which can cause increasing of waste and lead to pollution for centuries [8]. As an alternative, edible film or coating from biodegradable materials that can be degraded easily or might be consumed directly without producing waste [2]. It is produced from polysaccharides, proteins, lipids or combination of these materials. Besides of edibility and biodegradability, these materials are abundantly available and low cost. The functionality and performance of edible film as barrier to moisture and gases depend on film composition [9]. Chitosan is an amino polysaccharide derived from the N-deacetylation of chitin which exists naturally in exoskeleton of crustacean such as crab and shrimp [15]. It is whitish in color, odorless and tasteless. Chitosan is naturally abundant biopolymers that have excellent properties such as biodegradable, non-toxic, and bio-compatible [7]. Great attention of chitosan mostly due to its ability as film forming material in food industry as food packaging and preservative because it shows satisfactory gas barrier and mechanical properties [22].

Gelatin can be obtained by the controlled hydrolysis of the fibrous insoluble collagen present in the bones and skin generated as waste during animal slaughtering and processing [15]. Like chitosan, gelatin can form films and coating with good optical properties, adequate mechanical properties and excellent gas barrier properties [16]. Gelatin also has been found to contribute as major constituent in bio-plastic industry as food coating and preservative. Plasticizing agent are generally essential to overcome the weakness of film, mainly brittleness. By reducing intermolecular forces of polymer, plasticizer reduces the rigidity of film structure and increasing the mobility of biopolymeric chains, thus improving film's mechanical properties [18]. Glycerol has been proven to enhance film flexibility, reduce film puncture strength along with elasticity and also good water vapor barrier [20]. Besides, glycerol is mainly compatible for gelatin based film in improving mechanical properties and flexibility of the films [1]. Bergo et al. [4] found that the structure of gelatin film plasticized with glycerol showed no tendency of re-crystallization due to high stability and increasing of moisture in



the film. Fakhoury et al. [10] showed that manioc-gelatin film plasticized with sorbitol has stronger resistant through higher value of tensile strength and percent of elongation. Furthermore, it was found that amaranth flour films plasticized with glycerol give higher percent of elongation compared to films that used sorbitol [19]. The study by Isotton et al. [13] concluded that any plasticizers from the group of polyols were found to perfectly blend with starch based film. The structure was changed from malleable to brittle when using sucrose as plasticizer for starch based film [17]. The usage of sucrose as plasticizer to β -lactoglobulin film was found to increase the transition temperature T_g of the film which means that the suitability and bonding between film and plasticizer was not recommendable [6]. Many types of plasticizer have been used successfully in formulation of edible film for the purpose to improve their characteristics. Although numerous studies have been done on composite chitosan-gelatin film, this research was focused to investigate the effect of different type of plasticizer on physicochemical and mechanical properties of the film.

2. Materials and Methods

2.1. Materials

Chitosan and gelatin as raw materials were obtained from Merck. Glycerol (Merck Chemicals), sorbitol (Sigma Aldrich) and sucrose (System) acts as plasticizer.

2.2. Preparations of chitosan and gelatin films

The method was adopted from Hosseini et al. [12] with some modification. Chitosan (2% w/v) was prepared by dissolving chitosan in acetic acid (1% w/v) and stirred for four hours at temperature of 60°C. Gelatin (4% w/v) was prepared by dissolving gelatin in distilled water and stirred for half an hour at temperature of 40°C. Then, chitosan and gelatin film forming solution was mixed at ratio of 1:1, 1:2 and 2:1 and continuously stirred for another half an hour. Glycerol, sorbitol and sucrose were added to each film forming solution prior to casting on glass plate. Then it was dried in an oven at 55°C for 24 hours.

2.3. Mechanical Properties

Universal Tensile Machine (Tinius Olsen, H50KT) was used to measure tensile strength and percent elongation of the films. The test was carried out according to ASTM D-682 with cross head speed of 10 mm/min. Tensile strength was calculated by dividing the maximum load for the film to break cross-sectional area, and percent elongation by dividing film elongation at rupture to initial gauge length. Percent elongation is the ratio of extension to the length of the sample.

2.4. Physicochemical Properties

Physicochemical properties tested included moisture content, film thickness and color. Moisture content was carried out by drying each of the film in an oven at 105°C. The initial and final weights were recorded. Meanwhile film thickness was measured by using micrometer (Mitutoyo, Series 103-177) at five different points then the values were averaged. Color measurement was carried out by using chromameter where the measurement results were displayed in Hunter values of lightness, L^* , redness or greenness, a^* and yellowness or blueness, b^* .

3. Results and Discussions

3.1. Tensile strength

Figure 1 shows the effect of chitosan: gelatin ratio on tensile strength for different type of plasticizer. It is observed that the highest tensile strength was the film plasticized with glycerol compared to sorbitol and sucrose. Similar finding was obtained by Srinivasa et al. [18], where the chitosan/fatty acid films plasticized with glycerol had greater tensile strength than sorbitol and sucrose. Glycerol was miscible easily with chitosan and became more flexible compared to other plasticizers. This is also believed that due to molecular weight of glycerol is smaller than sorbitol and sucrose; it influences the mechanical properties of the film, thus giving more flexibility than other plasticizers. Al-Hassan and Norziah [1] found that the tensile strength for sago starch/fish gelatin films plasticized with sorbitol

had the highest tensile strength compared to glycerol. However, glycerol was found to be more effective compared to sorbitol when dealing with polysaccharides, protein and polysaccharide-protein based films because of higher mobility of polymer chains. Hence, the polysaccharides based film become more flexible and stretchable when using glycerol as plasticizer compared to sorbitol.

For films at ratio of chitosan: gelatin of 1:2, the tensile strength was the highest when plasticized with sorbitol compared to glycerol and sucrose. Chen et al. [6] reported that sorbitol plasticized films had slightly higher tensile strength than glycerol and polyethylene glycol (PEG) but increasing of plasticizer concentration, glycerol plasticized films showed higher tensile strength than sorbitol and PEG. From Figure 1, film plasticized with sucrose have the lowest tensile strength compared to other plasticizers. The usage of sucrose as plasticizer changed the film from malleable to brittle film [17]. Similar result was obtained by Al-Hassan and Norziah [1]. According to ratio of chitosan: gelatin, the highest tensile strength was recorded at 4.04 MPa for ratio 2:1. This shows that the higher the amount of chitosan, the higher the tensile strength of the film. The result was supported by Xu et al. [21] that found the tensile strength of composite film of chitosan and starch was increased by the increasing amount of chitosan leading to stronger films produced.

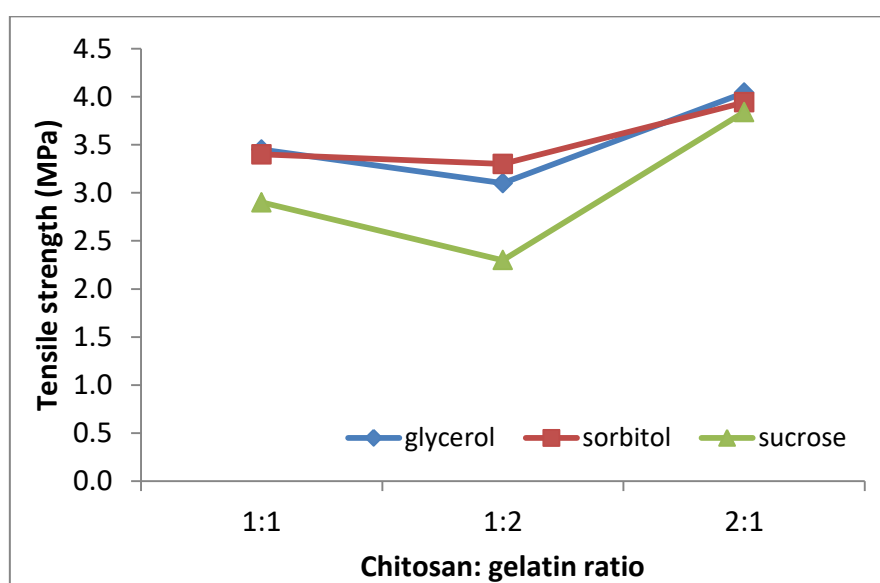


Figure 1. Effect of plasticizer on tensile strength for different chitosan:gelatin ratio.

3.2. Percent elongation

The effect of plasticizer on percent elongation for different chitosan:gelatin ratio is illustrated in Figure 2. It is shown that films plasticized with sorbitol had the highest percent elongation compared to glycerol and sucrose. Similar finding also found by Chen et.al [6], Arvanitoyannis et al. [3] and Srinivasa et al. [18] when comparing the percent elongation of films plasticized by few plasticizers such as glycerol, PEG and palmitic acid. Fakhoury et al. [10] reported that the sorbitol-plasticized films had about 20% higher percent elongation compared to glycerol-plasticized films. According to them, during the drying of films, water was evaporated and allowed the formation of starch network that bring the chains very near to each other, facilitating the formation of a denser matrix. However, several researchers had found that glycerol was the effective plasticizer which produced films with better percent elongation compared to sorbitol [5, 11]. The effectiveness of glycerol is most likely due to its small molecular weight, highest number of hydroxyl groups at the same concentration and hydrophilic nature which attracts water into the polymer matrix thus the effective level of plasticizer increases [11].

As shown in Figure 2, the highest percent elongation was obtained by glycerol plasticized film with ratio 2:1. While film plasticized with sucrose have the lowest percent elongation compared to sorbitol and glycerol. According to Fadini et al. [9], this is might be the conversion of amorphous sucrose to crystalline sucrose, resulting in brittle films that weakened and cracked. Although sucrose

plasticized film exhibit lower percent elongation than others for chitosan:gelatin ratio of 1:1 and 1:2, the use of sucrose can be a great alternative plasticizer since it exhibit higher tensile and percent elongation properties compared to films without any plasticizer [14].

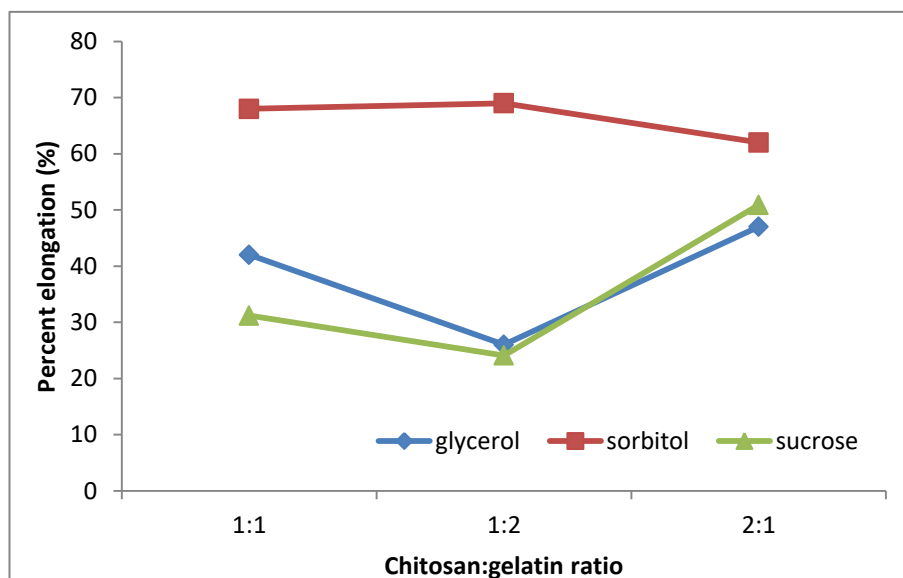


Figure 2. Effect of plasticizer on percent elongation for different chitosan:gelatin ratio.

3.3. Thickness

Table 1 shows the thickness, moisture content and color parameter for each of the film at different chitosan:gelatin ratio and plasticizer. Film plasticized with glycerol have the highest thickness which was 0.20 mm for ratio 1:1 followed by sorbitol and sucrose as shown in Table 1. This is because of hygroscopic nature of glycerol, the ability to retain moisture between its molecules resulted the film to be thicker than others. Fakhoury et al. [10] found that the thickness of chitosan-starch film was increased with the used of glycerol compared to sorbitol. Glycerol plasticized film produced thicker film because of its hydrophilic properties and higher protein content than other films [1]. Furthermore, films with ratio 1:2 have greater thickness than films with ratio 2:1. According to Hosseini et al. [12], biodegradable film produced with chitosan was found to decrease in water vapor permeability which led to thinner film produced. The results obtained were contradicted might be due to the addition of chitosan enhanced the cross-linking of gelatin and increase the free volume of the polymeric matrix.

3.4. Moisture content

Films with high chitosan content possess higher moisture content compared to films with high gelatin content as shown in Table 1. According to Al-Hassan & Norziah [1], the films produced from protein or polysaccharides material with chitosan is highly sensitive to moisture and exhibit poor water vapor resistance. Thus, the results obtained showed that the films with highest chitosan amount (ratio of 2:1) contain the highest moisture (27.87% for glycerol, 20.54% for sorbitol and 19.78% for sucrose). On the other hand, the films produced with chitosan-gelatin ratio of 1:2 shows the lowest amount of moisture content which are 19.78%, 17.24% and 17.65% for glycerol, sorbitol and sucrose, respectively. This shows that the lower the amount of chitosan, the lower the moisture content in the films. Moreover, glycerol plasticized films contains the most moisture content compared to sorbitol and sucrose. Bergo et al. [4] found that the used of glycerol among other plasticizers showed the greatest moisture content and increase significantly with the increasing amount of glycerol. Glycerol was identified having hygroscopic nature due to high likely to absorb moisture. Thus, it tends to retain water in the films than other plasticizer.

Table 1. Thickness, moisture content and color parameters for each films.

Plasticizer	Chitosan-Gelatin Ratio	Thickness (mm)	Moisture Content (%)	Color		
				L*	a*	b*
Glycerol	1:1	0.20	24.00	43.93	-0.46	1.42
	1:2	0.20	19.78	40.91	-0.44	1.42
	2:1	0.16	27.87	43.10	-0.90	3.35
Sorbitol	1:1	0.18	19.42	43.44	-0.43	1.48
	1:2	0.20	17.24	43.35	-0.41	1.69
	2:1	0.18	20.54	40.25	-0.82	2.46
Sucrose	1:1	0.16	19.23	42.79	-0.36	1.85
	1:2	0.19	17.65	40.65	-0.45	1.85
	2:1	0.14	19.78	43.07	-0.63	2.14

3.5. Color

Color of the films are expressed in term of L* (lightness or brightness), a* (redness or greenness) and b* (yellowness or blueness). From observation, overall films produced were slightly transparent. From Table 1, the yellowness of the films increases with the increasing amount of chitosan. The films produced with doubled amount of chitosan (ratio 2:1) shows higher lightness value than films with doubled amount of gelatin (ratio 1:2) except for films plasticized with sucrose. The values of a* and b* increase with the increasing amount of chitosan except for sucrose plasticized film. It indicates that, the higher the amount of chitosan in the film, the higher the tendency of films to be yellowish and greenish.

4. Conclusion

In summary, it can be concluded that the glycerol plasticized films have the highest tensile strength compared to sorbitol and sucrose at any ratio. The highest percent elongation was obtained by sorbitol plasticized films followed by glycerol and sucrose. Sucrose was found to make films from malleable to brittle despite of any ratio used. Due to hygroscopic nature, glycerol tends to promote higher moisture content for films containing double amount of chitosan and decrease the thickness. From observation, there has been no significant difference in colour for all films produced. Based on the value of L*, a* and b*, the higher amount of chitosan resulted the higher lightness, yellowness and greenness value.

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