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Study of Different Concentrations of Sorbitol as a Plasticizer in Producing Gelatin-Based Biodegradable Film from Chicken Claw Waste

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ABSTRACT

An environmentally friendly packaging material that can be used in place of synthetic, non-green plastic packaging is biodegradable film. Therefore, this study aims to determine the effect of sorbitol plasticizer concentrations on the characteristics of biodegradable film produced from chicken claw waste. The treatments applied during the production process consisted of three sorbitol concentrations, including 0.5, 1, and 1.5%, repeated thrice. Characterization of biodegradable film comprised thickness, water content, water solubility, film swelling, mechanical properties, pH, water vapor permeability, oxygen permeability, and degradation tests. Analysis of variance results showed that the sorbitol concentrations significantly influenced thickness value, but did not affect other characteristics. The best treatment was 0.5% sorbitol which generated thickness of 0.15 mm, WC of 13.97%, WS of 44.72%, swelling of 96.97%, tensile strength of 12.29 MPa, elongation of 22.23%, elasticity 58.53 Mpa, pH 7.5, WVP 9.26 g.m^{-1} pa^{-1} hour^{-1}, OP 1.52 g.m^{-1} pa^{-1}, and degradability for 14 days.

Keywords: Biodegradable film; chicken claw waste; gelatin; plasticizers; sorbitol

INTRODUCTION

Synthetic plastic is difficult to decompose, leading to environmental pollution and harmful effects on health when consumed. Plastic produced from petrochemicals cannot be broken down by microorganisms due to the low biodegradability. To carry out effective decomposition, microorganisms require special enzymes from an external source (Darni & Utami, 2010). The increasing use of plastic material for packaging is attributed to advantages, including lightness, durability, ease of shaping, anti-corrosion, good chemical resistance, electrical conductor insulation, and suitability for coloring addition (Mujiarto, 2005). Since this plastic type is unsafe, innovative technology is needed to develop environmentally friendly and consumable packing materials.

Biodegradable film is a material with thin, soft, and edible layer characteristics, appropriate for protecting or serving as packaging for eco-friendly products. Because of the ability to prevent the loss of water and air from products, as well as the mass transfer of solids into a material (Bustillos et al., 1994), biodegradable film is considered a new step and breakthrough to restore the healthy status of the earth. Plasticizers are added to reduce stiffness, increase elastic properties, and enhance the durability of biodegradable film produced from gelatin using chicken claw in the food packaging process.

Chicken claw, skin, and bones comprising a high content of unexploited collagen are less popular animal parts, often processed cheaply alongside other by-products such as feathers (Santana et al., 2020). The chemical content of chicken claw is 5.6% fat,

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65.9% water, 22.98% protein, 3.49% ash, and other component estimated at 2.03% (Purnomo, 1992). Chicken claw has the potential to be further developed due to containing high protein, specifically collagen, that by hydrolysis can be transformed into gelatin (Choe & Kim, 2018). The processed products from animal gelatin include biodegradable film (Khedri et al., 2021), It has good mechanical qualities and is frequently used in food packaging because of its permeability to gas and air (Etxabide et al., 2021) and a transparent nature (Ji et al., 2021; Wang et al., 2021).

Certain studies previously manufactured gelatin from chicken claw waste (Fatima et al., 2022; Ratna et al., 2023), but there have been no reports regarding biocomposite food packaging film produced with a gelatin/carboxymethyl cellulose (CMC)/sorbitol formulation. CMC is incorporated into biodegradable film to increase stability and compactness, while sorbitol addition enhances elasticity, preventing brittleness or easy cracking. Thus, the purpose of this work is to ascertain how the concentration of sorbitol plasticizer affects the properties of gelatin-based biodegradable film made from chicken claw waste. The characterization test examined swelling, thickness, water solubility (WS), pH, water content (WC), oxygen permeability (OP), water vapor permeability (WVP), mechanical properties, and degradability, which were expected to be improved in gelatin-based biodegradable film packaging.

METHODS

Materials

The study employed broiler chicken claw waste that was purchased from vendors in Aceh Besar and Banda Aceh, Indonesia, while the engaged equipment included a microwave, digital scale, hot plate, oven, and silicone mold. Additionally, German Emsure® NaOH pellets were purchased for analysis, alongside German Merck KGaA acetic acid (CH_3COOH), NaCl (technical), butterfly brand CMC (technical), sorbitol (technical), and distilled water.

Biodegradable Film Production Process

During the preparation of modified agar (Ratna et al., 2022; Ratna et al., 2023), fresh chicken claw was soaked in 0.2 molar NaOH for 48 hours and the solution was changed every 24 hours. This mixture was washed until pH approached neutral, then soaking was continued in 0.05 M acetic acid solution for 24 hours before washing and extraction with distilled water. The ratio of claw to distilled water was 1:5 (w/v) and the extraction process was performed over a period of 4 hours using a microwave with 180 W power, followed by drying at 50 °C until the maximum WC became 16% to generaté gelatin.

Biodegradable film production implemented a non-factorial Completely Randomized Design (CRD) featuring 0.5% (S1), 1% (S2), and 1.5% (S3) v/v sorbitol concentrations, which were repeated thrice, and other treatments included 2% (w/v) gelatin, 1% (w/v) CMC, as well as 200 mL distilled water. The gelatin and 1% CMC were dissolved in 60 mL and 80 mL of distilled water using a hot plate while stirring, respectively. The solution from both material was mixed, distilled water was added until reaching 200 mL, and reheating was conducted for thirty minutes at 60 °C on a heated plate while stirring continuously. This was cooled for 5 minutes and sorbitol was introduced at concentrations corresponding with the

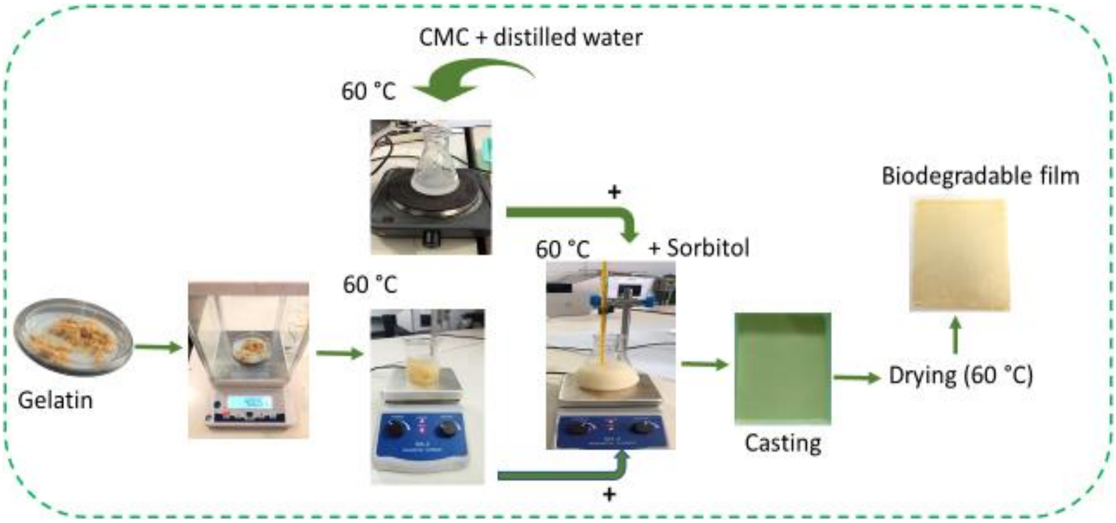

Diagram 1. Study scheme

treatments before being transferred into a silicone mold measuring 20.5 cm x 18.5 cm. Drying was performed using an oven at 60 °C for 20 hours, after which biodegradable film produced was removed from the mold.

Characterization of Biodegradable Film

Thickness

After the film was taken out of the mold, its thickness was measured at nine different random locations using a screw micrometer. Subsequently, an average of the measurements was calculated as thickness of biodegradable film produced in this study (Ratna et al., 2022, Ratna et al., 2024; Ratna et al., 2023; Ratna et al., 2023).

Water Solubility (WS) and Water Content (WC)

Empty 50 mL bottles were coded and the initial weight W1 was measured, then the 30 mm x 30 mm film was added and weighed as W2. Heating was performed in the oven at 105 °C for 24 hours and dry weight was calculated as W3, then 15 mL of distilled water was introduced into the bottles containing film and stirred before stagnating for 24 hours and discarding water component. Drying was conducted again at 105 °C for 24 hours, followed by weighing to obtain W4, WC, and WS using Equations 3 and 4 (Ji et al., 2021).

$$\text{WC (\%)} = \frac{W2-W3}{W2-W1} \times 100\% \qquad (1)$$

$$\text{WS (\%)} = \frac{W3-W4}{W3-W1} \times 100\% \qquad (2)$$

Swelling

Film produced with dimensions of 30 mm x 30 mm was evaluated for dry weight (Wi), then soaked in distilled water over a period of 10 minutes and removed. Using filter paper, the remaining water on the film was removed, and the wet weight (Wf) was calculated, while the degree of swelling was calculated using Equation 5 (Syahida et al., 2020).

$$\text{Swelling (\%)} = \frac{Wf-Wi}{Wi} \times 100\% \qquad (3)$$

Mechanical Properties

Biodegradable film specimens were prepared and examined following ASTM method D 638-03 using Universal Testing Machines (Hung Ta Type HT-8503, Taiwan). Furthermore, the mechanical characteristics of the nanogelatin film were assessed, including its tensile strength, elongation, and Young's modulus.

pH

The 30 mm x 30 mm film was soaked in distilled water for 10 minutes, then removed and pH was measured using pH meter (Ratna et al., 2022).

Water Vapor Permeability (WVP)

The 9 mL bottles were filled with 6 mL of distilled water and covered with biodegradable film measuring 30 mm x 30 mm. These were weighed and positioned in a desiccator at room temperature, then new weight was measured every 24 hours for 3 days. WVP of film was calculated using Formula 6, which was the modification of a previously described procedure (Ji et al., 2021; Ratna et al., 2022).

$$\text{WVP } (g.m^{-1}.Pa^{-1}.h^{-1}) = \frac{\Delta m \times d}{A \times t \times \Delta P} \qquad (4)$$

Where d is film thickness (mm), m represents the mass loss (g), A denotes the area of the bottle mouth (m^2), t is the measurement time interval (h), and p signifies the partial vapor pressure (4247 Pa) difference between the two sides of film at room temperature and relative humidity of 100%.

Oxygen Permeability (OP)

OP measurement was conducted based on the modification of a previous procedure (Yadav et al., 2020; Ratna et al., 2022), where biodegradable film was cut into 30 mm x 30 mm. The bottles used were 9 mL in size and gelatin film was placed over the mouths, and the starting weight was recorded. These were transferred into a desiccator, then weighed every 24 hours for 3 days, while Oxygen Permeability Transmission Rate (OPTR) and oxygen permeability were calculated using Equations 7 and 8 (Yadav et al., 2020).

$$\text{OPTR} = \frac{Slop}{Film\ area} \qquad (5)$$

$$\text{OP} = \frac{OPTR \times L}{\Delta P} \qquad (6)$$

P = Partial vapor pressure differential between dry atmosphere and pure water (0.02308 atm/1.013×10^5 Pa at 25 °C), and L represents the average biodegradable film thickness. Meanwhile, slop is the slope value of the line on a linear regression graph with the X axis as time (per hour) and the Y axis being the weight (g) of biodegradable film sample.

Degradation Test

Degradation test was performed by cutting biodegradable film into individual sizes measuring 30 mm × 30 mm. Biodegradable film sample was planted without covering (aerobically) to promote direct contact with bacteria and fungi present in the soil, a process also known as soil burial test (Haryati et al., 2017). Observations were conducted every 3 days to monitor the visual changes that occurred until the sample experienced degradation.

Statistic Analysis

The test result data obtained were the mean ± standard deviation (SD), examined using One-way Analysis of variance (ANOVA). This was followed by the Duncan test at a significance level of $p < 0.05$, and the entire collected data were analyzed using SPSS ver.24.

RESULTS AND DISCUSSION

Thickness

Thickness of biodegradable film can influence the tensile mechanical properties, elongation rate, WVP, and OP. The results of thickness testing conducted on biodegradable film samples showed different values, as presented in Figure 1.

Figure 1 shows that the application of greater sorbitol concentration in the current study leads to the production of thicker biodegradable film. This product was similar to the type reported by Ratna et al. (2022), where greater sorbitol concentration generated thicker sample. Therefore, a sorbitol concentration of 0.5% produced the smallest thickness, and 1.5% generated films with the largest thickness ranging from 0.15 – 0.17 mm. Based on the results, all treatments with different sorbitol concentrations still produced thickness adhering to the Japan International Standard with a maximum value of 0.25 mm. The sorbitol concentrations were significantly different *p<0.05 from thickness value of biodegradable film. Based on the results of the Duncan Advanced Test, sample S1 was found to be insignificantly different from S2 but it varied from S3, while S2 was insignificantly different from S3.

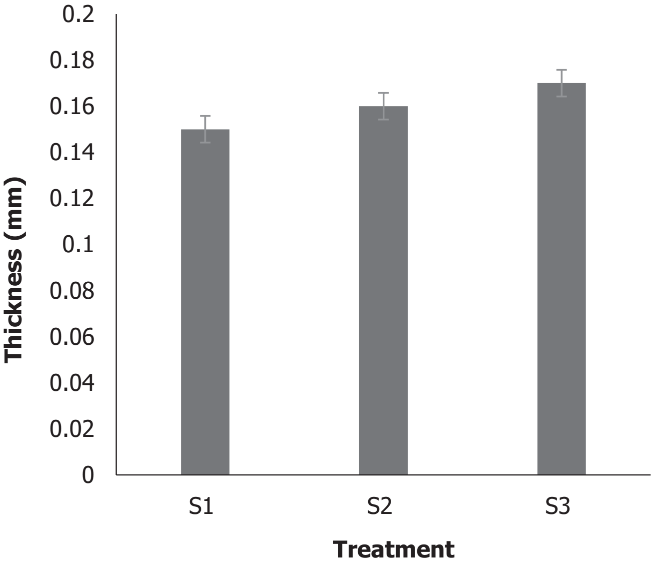

Figure 1. The average value of biodegradable film thickness for each sorbitol concentration treatment

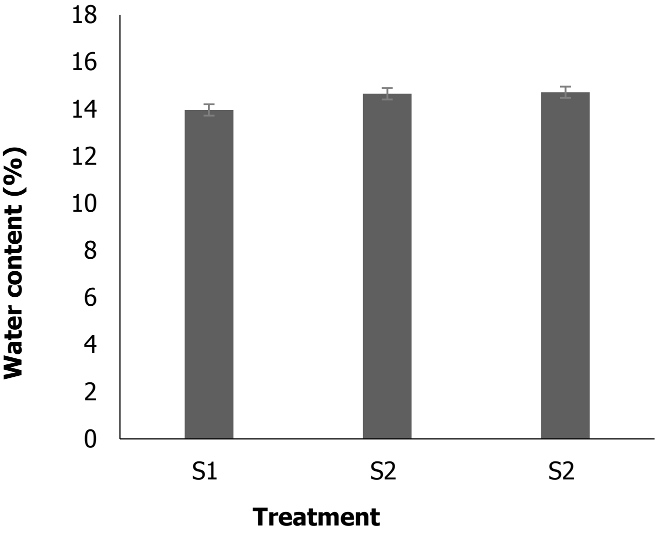

Figure 2. The average WC value of biodegradable film at each sorbitol concentration treatment

Water Content (WC)

WC is the amount of water present in biodegradable film, therefore, it greatly influences the shelf life of the packaging material. The particular relationship existing between WC and sorbitol concentration added during the experiment is shown in Figure 2.

The purpose of WC testing conducted was to determine the total WC present in the resulting biodegradable film packaging. According to a previous study (Setiani et al., 2013), a packaging containing high WC would be easily attacked by microorganisms, specifically the type comprising high protein, but those with low WC could be more resistant to attacks. Biodegradable film data in Figure 5 showed that treatment with sorbitol concentrations of 0.5%, 1%, and 1.5% generated the lowest WC value at 13.97%, followed by 14.65%, and the highest WC at 14.72%, respectively. The sorbitol concentrations were significantly different at p-value >0.05 compared to WC value of biodegradable film.

Water Solubility (WS)

WS shows the ability of biodegradable film to absorb water and dissolve, leading to easy decomposition when

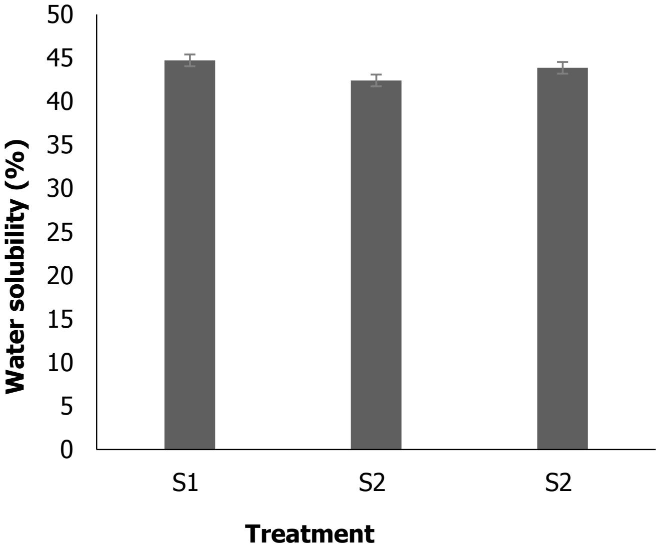

Figure 3. WS of biodegradable film

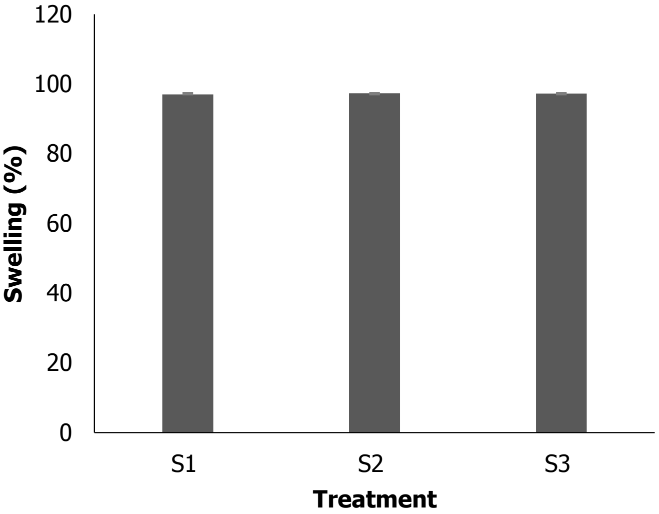

Figure 4. Average swelling value of biodegradable film for each sorbitol concentration treatment

thrown directly into the environment. Furthermore, the entire WS values of biodegradable film produced in this study are respectively presented in Figure 3.

Solubility of biodegradable film in water can determine the appropriate application for food packaging (Dogaru et al., 2021), while the hydrophilic and hygroscopic nature affects film solubility. Buffer plasticizers interact with film polymer which tends to increase the space between chains, supporting the transfer of water into the internal compartment, thereby increasing solubility (Ahmadi et al., 2021). Biodegradable film with low solubility is often applied to food as packaging, but the higher WS level will lead to a more rapid degradation process. According to Figure 4, 0.5%, 1.5%, and 1% sorbitol concentration treatments generated the highest WS results at 44.72%, followed by 43.86%, and the lowest was 42.41%, respectively. The sorbitol concentrations showed no significant difference at $p>0.05$ compared to WS value of biodegradable film.

Swelling

The development test was conducted to determine the percentage of swelling that occurred in the produced film due to water presence. Swelling tends to be caused by materials possessing hydrophilic properties, namely those capable of binding water (Yadav et al., 2020). The effect of sorbitol concentration on the development of biodegradable film for each sample is presented in Figure 4. The lowest swelling with a value of 96.97%, followed by 97.26%, and the highest at 97.37% was initiated by 0.5%, 1.5%, and 1% sorbitol concentration treatments, respectively. Swelling increases with higher water absorption capacity, and vice versa, while the occurrence of water absorption at a high speed leads to swelling (Yadav et al., 2020). The sorbitol concentration did not show a significant difference at $p>0.05$ in the development of biodegradable film.

Mechanical Properties

The ability of a film to sustain maximal stress before rupturing is known as its tensile strength, and the values obtained among biodegradable films in this study vary as shown in Figure 5. Biodegradable film is expected to have high tensile strength values in order to be used as quality packaging for a product, and sorbitol addition can increase tensile strength. Plasticizers affect the internal hydrogen bonds of film molecules to weaken the attraction of intermolecular hydrogen bonds in the polymer chain, thereby reducing the breaking strength (Ratna et al., 2021). The highest tensile strength value of 14.29 Mpa and lowest measuring 12.29 Mpa were obtained at sorbitol concentrations of 1.5% and 0.5%, respectively. Japanese Industrial Standards (JIS) recommended a minimum tensile strength of 0.39 Mpa and Indonesian National Standard (SNI) suggested a minimum of 1-10 Mpa. Therefore, the three treatments used in this study produced tensile strength values meeting JIS and SNI recommendations. According to a previous investigation (Setiawan & Aulia, 2017), the high tensile strength value obtained showed that biodegradable film was suitable for use as environmentally friendly packaging. Sorbitol concentrations presented a significant difference at $*p \leq 0.05$ for tensile strength value of biodegradable film. The Duncan Advanced Test results showed that treatment S1 was significantly

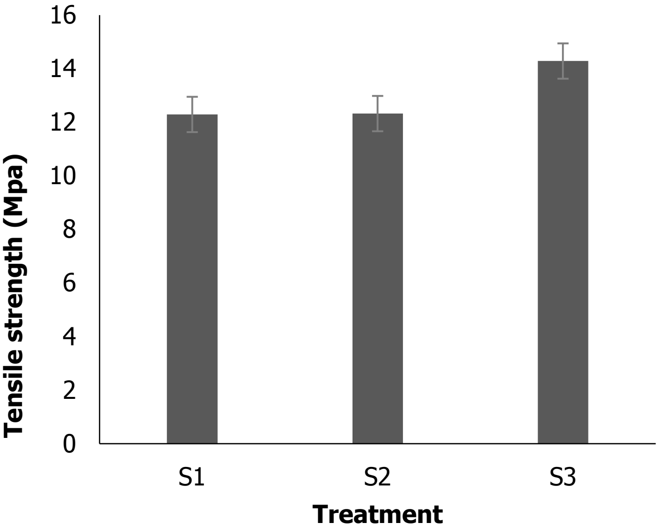

Figure 5. Average tensile strength value of biodegradable film for each sorbitol concentration treatment

different from S3, while S2 varied from S3, and S1 was not significantly different from S2.

Elongation test aims to determine film ability to elongate, which is calculated until the material breaks, and the results can be seen in Figure 6. According to JIS, elongation value below 10% for biodegradable film is not good, but those above 50% are very good. Based on SNI, the characteristics of good biodegradable film range from a minimum of 20 – 21%. Elongation percentage in this study was different for each treatment, namely ranging from 20.13% to 25.41%. The applied sorbitol concentration of 1.5% generated the lowest value and 1% produced the highest elongation value. Treatments that met these two standards were 0.5% and 1%, while sorbitol concentration showed a significant difference at $*p \leq 0.05$ for film elongation value. The results of the Duncan Advanced Test showed significant differences between treatments S1 and S3 as well as S2 and S3, but S1 and S2 had no significant differences. Elongation value has an inverse relationship with tensile strength, thereby implying that a greater elongation value leads to smaller tensile strength.

Elasticity is a testing procedure that determines how resistant film material is to experiencing strain against the effort of the material to achieve the original shape after distortion by vertical external stress. The results of the relationship between the effect of sorbitol addition and elasticity is presented in Figure 7. The addition of sorbitol tends to increase film elasticity, thereby producing film that is not brittle or easily broken. According to a previous study (Bustillos et al., 1994), elongation percentage value exceeding 50% represented a good film category. Film is categorized as bad when the percentage value of film length is less than 10%, and Figure 6 shows various elongation results. The data obtained was that the 0.5%, 1%, and 1.5% sorbitol concentrations produced film reaching elasticity levels of 58.53%, 46.65%, and 76.10%, respectively. S1, S2, and S3 generated good elasticity values, while sorbitol concentrations presented a significant difference of $*p \leq 0.05$ for elasticity values of biodegradable film. The Duncan Advanced Test results

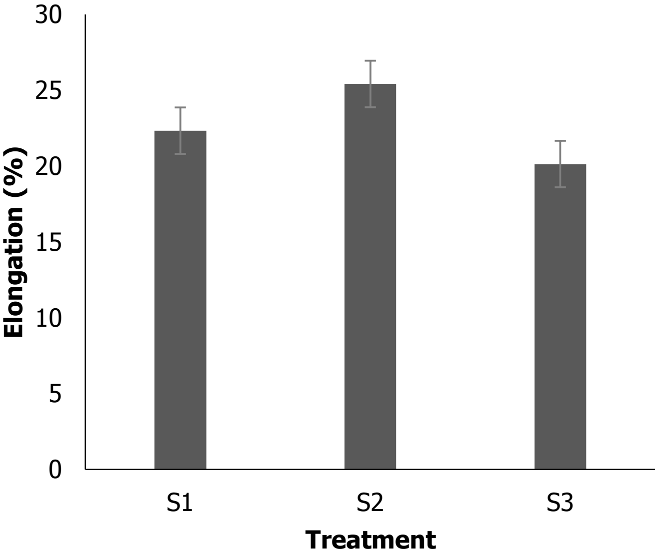

Figure 6. Average elongation value of biodegradable film for each sorbitol concentration treatment

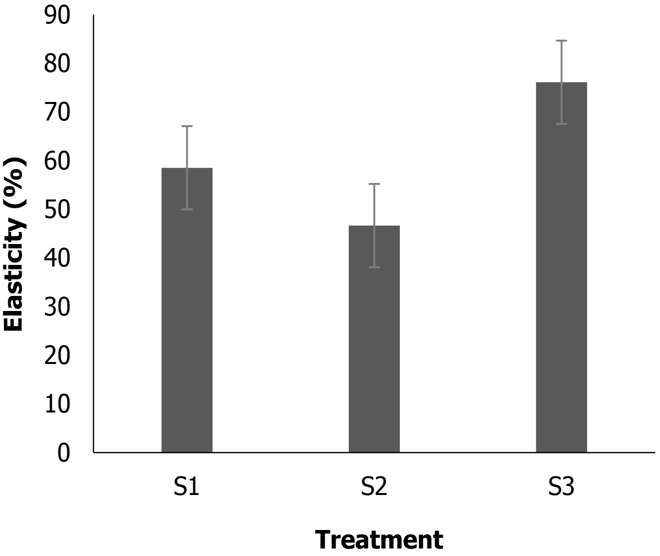

Figure 7. The average value of biodegradable film elasticity for each sorbitol concentration treatment

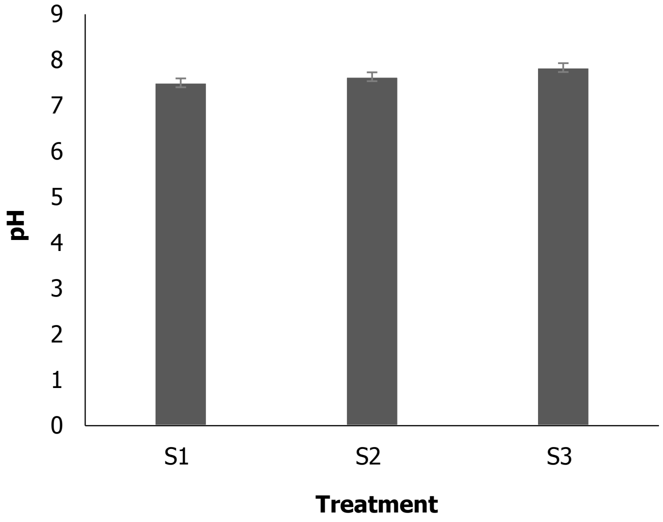

Figure 8. Average pH value of biodegradable film for each sorbitol concentration treatment.

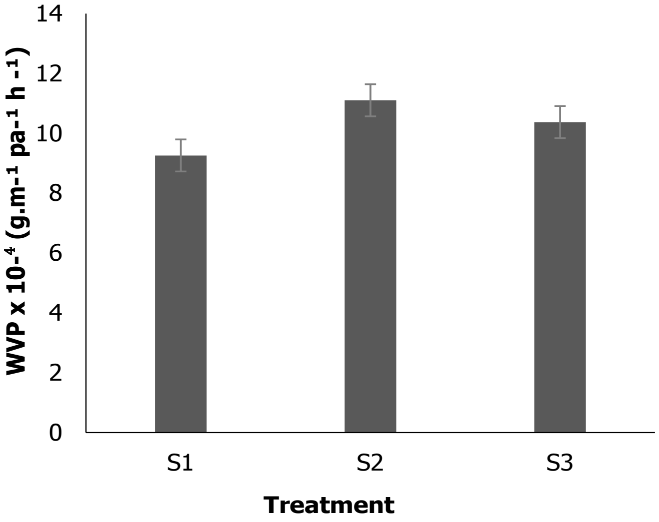

Figure 9. Average WVP value of biodegradable film for each sorbitol concentration.

showed real differences between S1, S2, and S3, but S1 and S2 were not significantly different.

pH

pH testing was performed to measure the acidity level of biodegradable filmproduced using gelatin obtained from chicken claw. The relationship between variations in sorbitol and pH of biodegradable film is presented in Figure 9, with the three treatments initiating pH ranges of 7.5 to 7.8. These values were similar to pH reported by Ratna et al. (2022) who conducted investigation on gelatin-based edible film with a glycerol concentration of 1%, which produced a pH of 6. All sorbitol concentration treatments produced biodegradable film with neutral pH, thereby promoting safety for use as food and edible packaging. However, the resulting pH changes observed were caused by the length of the gelatin dissolution process which also varied. ANOVA analysis showed that there was no significant effect of the sorbitol concentration treatment on pH value of biodegradable film with *p*-value >0.05.

Water Vapor Permeability (WVP)

WVP testing or water vapor transmission rate is a procedure to determine the transfer rate of water vapor that penetrates film to the environment at a certain temperature and humidity (Ratna et al., 2019; Ratna et al., 2021). The relationship between the influence of sorbitol concentration and WVP is presented in Figure 9. Based on a previous study (Anandito et al., 2012), when the rate of water vapor transfer from material packaged using biodegradable film appeared small, the shelf life of the material would be maintained. According to Figure 12, treatment with a sorbitol concentration of 0.5% produced good WVP values at 9.26 gm^{-1} pa^{-1} h^{-1}, while 1.5% generated 10.38 $g.m^{-1}$ pa^{-1} h^{-1} and 1% initiated the highest WVP value at 11.10 $g.m^{-1}$ pa^{-1} h^{-1}. Sorbitol concentration showed no significant difference at *p*>0.05 for pH value of biodegradable film.

Oxygen Permeability (OP)

The process known as OP is used to assess a bio-based film's ability to shield food items from lipid oxidation, volatile scents, moisture and oxygen transfer, and taste degradation. The relationship

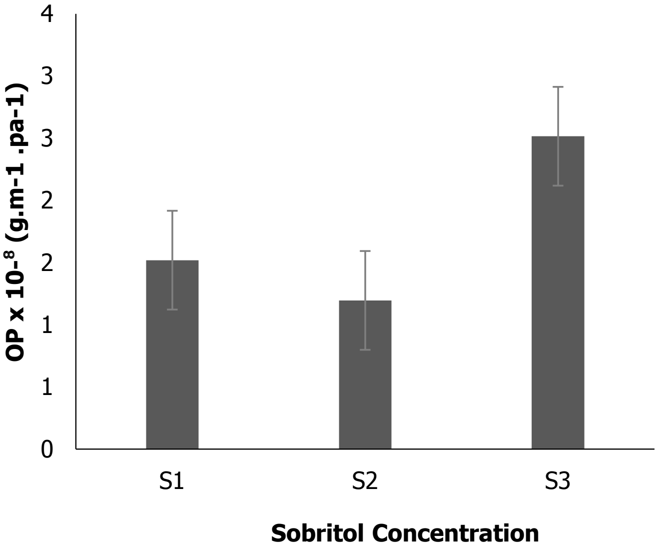

Figure 10. The average value of OP of biodegradable film for each sorbitol concentration

between OP and the effect of sorbitol concentration is presented in Figure 10. OP influences several changes in the quality of food packaging including oxidative degradation of food components, color changes in the packaging, and microbial growth. A low OP value can reduce the potential of damaging the material to be packaged, therefore, a high OP leads to poor packaging (Wongphan et al., 2022). The study data showed OP values of 1.20 g.m^{-1} .pa^{-1}. d^{-1}, 2.52 g.m^{-1} .pa $^{-1}$, and 1.52 g.m^{-1} .pa^{-1}.d^{-1} obtained in the 1%, 0.5%, and 1.5% sorbitol concentration treatments, respectively. Sorbitol concentration showed that there was a significant difference with $*p \leq 0.05$ for OP values of biodegradable film. The Duncan Advanced Test results showed real differences between S1 and S3 as well as S2 and S3, but S1 and S2 were not significantly different.

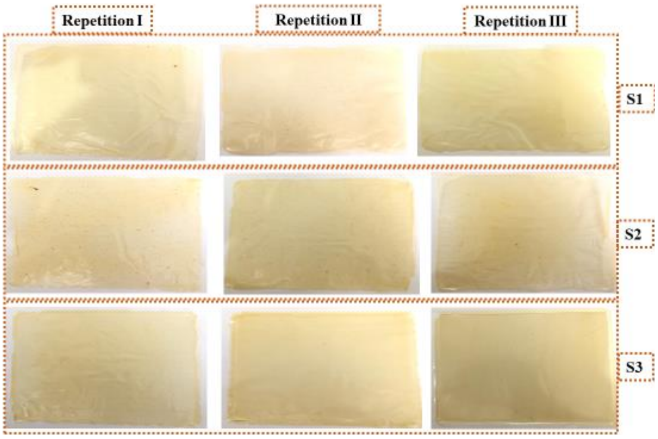

Figure 11. Gelatin-based biodegradable film from chicken claw waste

Degradation

The level of biodegradable film resistance can be determined through biodegradability testing to identify the effect of decomposing microorganisms, soil moisture, temperature, and physicochemical factors found in the soil (Zulferiyenni et al., 2014). This test is also known as the soil burial technique, often conducted by controlling the microorganisms to facilitate the decomposition process (Subowo & Pujiastuti, 2003), while the structures of biodegradable film are shown in Figure 11.

During degradation test, each sample was planted using soil gathered in a different container, which was observed for 14 days until film was fully decomposed. The decomposition process was also influenced by microorganisms along with moist soil conditions, and degradation test was carried out by visual observation. In a maximum time of 60 days according to SNI, film experiences changes in the material until decomposition.

Sample changes started occurring with variations in sorbitol concentrations of 1% and 1.5% during observations until day 3, as several holes were found in certain parts of film. On day 6, the surface of all variations of biodegradable film decreased as the days increased, which could be observed from the damage experienced with sorbitol concentration of 0.5%. The occurrence of damage showed the influence of soil microorganisms on the degradation process, and on day 12, the sample with a sorbitol concentration of 1.5% was decomposed. On day 14, all biodegradable films had a texture similar to the soil, showing that the samples were completely decomposed. Based on visual observations, there was occurrence of significant degradation, which was characterized by visible damages

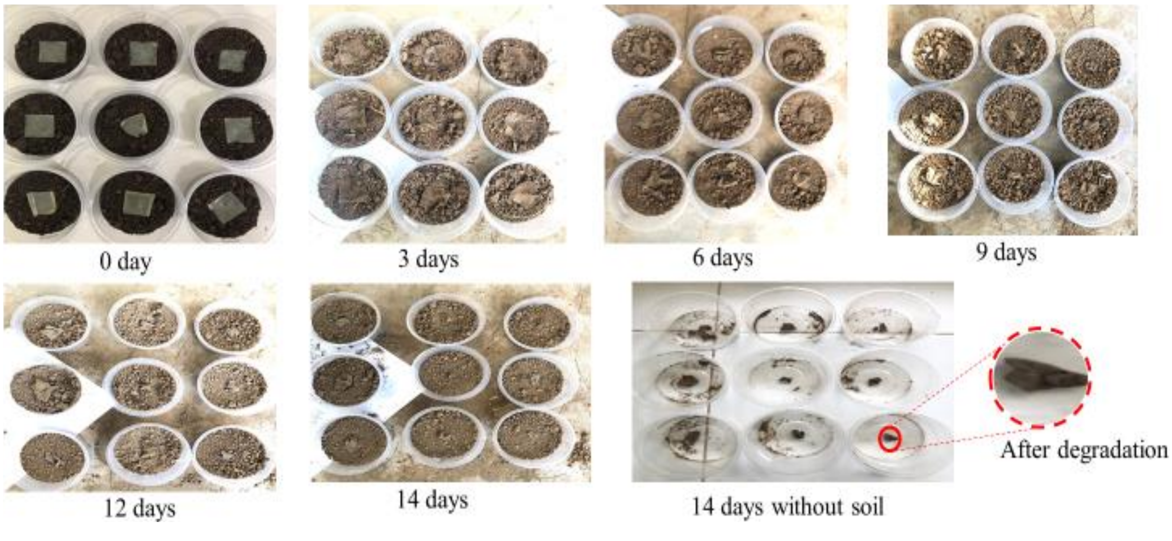

Figure 12. Changes in biodegradable film during degradation process

detected. Degradation test results were consistent with a previous study (Apriyani, 2015) where all film samples tested for degradation were decomposed and insignificant differences were found. This observation is due to the ability of microorganisms in the soil to break down film, but the microorganisms cannot decompose synthetic plastics (Latief, 2001).

CONCLUSION

In conclusion, the results showed that increasing sorbitol concentrations enhanced biodegradable film thickness, ranging from 0.15 – 0.17 mm, along with WC of 13.97 – 14.72%. WS described as film ability to dissolve in water showed values between 42.41 – 44.72%, while swelling ranged from 96.97 – 97.37%. In general, mechanical properties of biodegradable film were found to improve with increasing sorbitol concentrations. Smaller water vapor and OP values led to better barrier properties against water vapor and oxygen. In this study, higher sorbitol concentrations generated greater water vapor and OP values, while biodegradable film produced from chicken claw gelatin could decompose naturally.

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CONFLICT OF INTEREST

The authors declare no conflict of interest with other parties.

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