

Characteristics of Canna Starch-based Edible Film Produced using Various Types of Plasticizers and Their Application on Tomatoes

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Abstract

Currently, eco-friendly postharvest treatments are required to fulfill consumers' demands and extend fresh agricultural commodities' shelf life. Starch-based coatings offer promising potential for this application due to their desirable properties. The objectives of this research were to investigate the effect of various canna starch concentrations and plasticizer types on the characteristics of edible film and to evaluate their effectiveness in coating tomatoes applied by spraying and fogging methods. Results indicated that starch concentration significantly influenced moisture content, thickness, and water vapor transmission rate. The plasticizer type affected lightness, water solubility, and optical properties. The interaction between starch concentration and plasticizer type significantly impacted moisture content. Based on these findings, a coating formulation of 1 % starch concentration with sorbitol as a plasticizer was selected. The fogging application of this coating resulted in a uniform coverage on the tomato surface. These results highlight the potential of canna starch-based edible films as a sustainable and effective postharvest treatment for fresh agricultural commodities.

Keywords: Canna starch, Edible film, Coating, Tomatoes, Quality

Introduction

Increasing consumer awareness of the health benefits of food has led to a surge in the consumption of fresh produce, such as fruits and vegetables. Therefore, appropriate postharvest treatments are required to maintain the quality of fresh agricultural products for longer shelf life to meet consumers' expectations. The common procedures to maintain freshness includes low-temperature storage, controlled atmosphere storage, modified atmosphere packaging, and the application of

biodegradable coating. Coating, which covers a thin layer of materials on fruit surfaces, is a common practice for maintaining the quality and extending the shelf life of the fruits by serving as mass transfer barriers to moisture, oxygen, and carbon dioxide to the surrounding atmosphere [1]. Up to now, a considerable amount of research has already reported the effectiveness of the edible coating application in maintaining the quality of plums [2], tomatoes [3], strawberry [4], banana [5],

grapes [6], and chili [7]. The studies showed that the coating contributed to the retention of vitamin C, color, texture, freshness, less weight loss, and maintenance of consumers' acceptance. Besides, the edible coating is also useful in preserving the quality of fresh-cut fruit such as pear [8], papaya [9], kiwi [10] and preventing browning on mangoes [11] and apples [12].

Various bio-based polymers are abundantly available from plant and marine products for producing edible films. The edible film is mainly composed of polysaccharides, proteins, or lipids [1]. Among them, polysaccharides are the most common type of materials used for the development of edible coating [13]. Starch edible films are widely applied in practice due to several advantages such as film-forming ability, acceptable gas barrier, odorless, colorless, non-toxicity, biocompatibility, low cost, abundantly available, and environmentally friendly [14]. As the main material of the edible coating, starch can be obtained from rice [15] or cassava [16]. Aside from that, various other types of starch have not been widely utilized such as cassava bagasse starch [17], jicama starch [18], mung bean starch [19], arrowroot starch [20] and konjac glucomannan [21], are also could be developed as resources for edible film or coating.

In tropical countries, canna (*Canna discolor* L.) is abundantly available due to its ability to grow on marginal land and its low cultivation costs. Canna starch contains approximately 60 % amylopectin, 33 % amylose, 0.22 % ash, and 0.31 % crude fiber [22], making it a promising material for use as a coating on fruits [23]. Moreover, starch-based edible films are known for their favorable quality attributes and functionality, making canna starch a potential candidate for further development in this field [24]. However, previous studies reported that edible film made from starch shows weaknesses when applied as food packaging materials, including the relatively low water-barrier properties and unsatisfactory mechanical properties [25]. To overcome these limitations, several treatments are recommended including blending with other materials [23,26], incorporating nanoparticles or "nanosystem" [27,28] and adding of plasticizers [29-31]. Lipid has been used to improve the moisture resistance [32] and combined with protein can be used to enhance the water vapor barrier and elongation of the starch edible films [33]. The "nanosystem" being

applied includes nanoemulsions, polymeric nanoparticles, nanofibers, solid lipid nanoparticles, nanostructured lipid carriers, nanotubes, nanocrystals, nanofibers, or mixtures of organic and inorganic nano-sized components. The "nanosystem" enhances the properties of the biodegradable coatings by improving gas (O₂ and CO₂) and water vapor permeability, mechanical strength and optical attributes, and antioxidant/ antimicrobial activity [28].

Besides nanotechnical approaches, an addition of plasticizers, such as glycerol, sorbitol, and polyethylene glycol (PEG), in the solution of edible coating/film also contributes to improved water solubility and mechanical properties of the products. Compared to sorbitol, glycerol showed a better effect on improving the puncture strength and elongation [34]. The addition of glycerol to the edible film solution from potato starch resulted in better flexibility of the film compared to the other treatments [35]. On the other hand, sorbitol as plasticizer can enhance water vapor barrier and tensile strength compared to other plasticizers that commonly used in edible coating formulation [36], whereas PEG can increase optical transparency, lowest water solubility and have good thermal stability [31]. Glycerol used for edible film can improve moisture content of film and water solubility. The use of 3 different plasticizers (glycerol, sorbitol, and PEG) in the making of the edible film affects the characteristics and barrier performance [30].

In addition to applying an appropriate plasticizer, the method of coating application on fruit surfaces also impacts the quality maintenance of the treated fruits. Common practices of coating applications on fruit surfaces are dipping, brushing, or spraying [37]. The previous research has been applied spray technology edible coating based on xanthan gum-based onto food products through a nozzle [38]. For better results, it was recommended that the application of the multi-layer edible coating should be sprayed with nano-sized particles. Due to the high cost associated with nanoparticle applications, a fogging method using an electric fogger presents a viable alternative for applying edible coating solution. This method produces smaller particles sizes compared to the conventional sprayers, potentially enhancing the coating's effectiveness. Therefore, this research aimed to provide valuable insights into the fogging method as a cost-effective

alternative to nano-system applications. Specifically, the objectives of this research were to investigate the effect of various canna starch concentrations and plasticizer types on the characteristics of edible film and to evaluate their effectiveness in coating tomatoes using spraying and fogging application methods.

Materials and methods

Materials

Canna starch used in this study was obtained from CV. Progress Jogja (Yogyakarta). It contains 25.81 % amylose and 60.42 % of amylopectin. Plasticizers used include glycerol, sorbitol, and polyethylene glycol 400 (food grade), provided by a chemical distributor "Prima Kimia". Tomatoes variety "Servo" were harvested from the farm in Purwokerto. Fresh fruits with peduncle were transported in a box to the laboratory. Tomatoes samples used weighed about 60 g, diameter of ± 5 cm, and light red peel. The fruits were rubbed with a dry and soft cloth and then kept in an individual tray. This research was conducted using a randomized design with 3 replications for each sample.

The coating materials were the mixture of the canna starch solution at 1.0 % (G1), 1.5 % (G2), and 2.0 % (G3) with 3 types of plasticizers: Sorbitol (P1), glycerol (P2), and polyethylene glycol (PEG) 400 (P3). The observed parameters were moisture content, thickness, optical properties, lightness, water solubility, and water vapor transmission rate. Based on the initial investigation, the treatment that produced the best characteristic of edible film was chosen. Afterward, the solution was applied to tomatoes by spraying and fogging methods. Surface morphology and image analysis of the tomato surfaces were investigated.

Preparation of edible films

Canna starch solutions of 1.0, 1.5 and 2.0 % (w/v) were prepared, with different types of plasticizers added at 30 % (v/w) of the starch weight. During stirring, distilled water was dropwise added to reach 180 mL. Subsequently, 1 % (v/v) acetic acid was incorporated into the solution. The mixture solution was then homogenized while being heated on a hotplate until a visible gel formed. An 80 mL portion of the gel was cast onto a glass mold (10×20 cm) and placed in the cabinet dryer (Raja Pengerang, Indonesia) for approximately 10 h at a temperature of 50 - 55 °C. After drying, the edible

film was cooled to room temperature before being removed from the mold and stored in a desiccator.

Application of coating on the tomato

Coating by spraying method

The coating solution was prepared in the container connected to a nozzle with a diameter of 1 mm. The spraying process was conducted with a sprayer positioned 60 cm from the sample, and the pump pressure (Morris MOC9, Indonesia) was set at 3 bar. The sample was hung and rotated slowly to ensure the homogeneity of the coating process. Afterward, the coated tomatoes were dried using a standing fan (Panasonic, Indonesia) for 5 minutes. The treated samples were then collected for further analysis.

Coating by fogging method

The application of the coating was conducted using an electric fogger. The coating solution was placed in a tank and flowed to the fogger when the machine was turned on. The fogger produced a fine mist inside a chamber where the tomato was suspended at the center. This process was conducted for 30 s. Afterward, the tomato was moved to the next chamber for the drying process, where warm air was blown into the chamber for about 1 min. Once dried, the coated tomatoes were prepared for the analysis. The coating and drying process was conducted individually for each fruit.

Analyses of edible film

Moisture content

The moisture content of edible film was analyzed with gravimetry methods which refers to AOAC [39]. After the initial weight was measured, the sample was dried in a drying oven (Mettler UN 55, Germany) at 105 °C for 24 h. The sample was removed and placed in a desiccator for 1 h before it was checked again for the final weight using an analytical balance (Sartorius MSA225S, Germany).

$$\text{Moisture content (\%)} = ((W_w - W_d) / W_w) \times 100 \quad (1)$$

where, W_w was the initial weight of the film and W_d was the constant weight of film after drying.

Thickness

The thickness of the edible film was measured by using the micrometer (Krisbow, Indonesia) with an accuracy of 0.001 mm at 5 different points [39].

Opacity

The determination of the opacity was conducted using a UV-Vis spectrophotometer (Shimadzu, Japan) [33]. Film samples measuring 4×0.9 cm were placed in cuvette, and their absorbance at a wavelength of 550 nm was recorded. The opacity of the sample was then calculated based on the following equation:

$$T = A_{550}/T \quad (2)$$

where A_{550} is the absorbance at 550 nm, T is the thickness of the film (mm).

Lightness

The lightness of the film was represented by the L^* value, measured by Color Reader (Konica Minolta CR-10, Japan). The average of L^* values was obtained from the measurement at 5 different spots of the film [19].

Water solubility

The samples were cut into 2×2 cm² size and dried in the oven (Memmert UN 55, Germany) at 105 °C for 24 h. After drying, samples were placed in a desiccator and weighed using an analytical balance (Sartorius MSA225S, Germany). Subsequently, the sample was immersed in 10 mL distilled water for 6 h. The wet sample was dried again in the oven at 105 °C for 24 h, and then placed in the desiccator [25]. Samples were weighed to determine the final dry mass. Water solubility was measured using the following equation:

$$\text{Water solubility (\%)} = ((W_1 - W_2)/ W_1) \times 100 \quad (3)$$

where W_1 was the initial weight of the film and W_2 was the weight of film after solubilization.

Water vapor transmission rate (WVTR)

The edible film samples were cut into a round shape to fit the top of a test cup (6 cm in diameter). The film was placed over the test cup and sealed tightly

around the edges using melted paraffin. The test cup was then placed inside a sealed glass jar to ensure controlled conditions during the experiment. The test cup was weighed every hour for over a period of 7 h. The changes in weight over time were recorded, and the slope of the weight change was determined using linear regression analysis [35]. The WVTR was calculated using the following equation:

$$\text{WVTR (g/h/m}^2\text{)} = ((W)/(A \times T)) \quad (4)$$

where: W is change in weight (g); A is surface area of the film, and T is time (h).

Surface morphology of tomato

To observe the surface morphology of the coating applied to the tomato, Scanning Electron Microscopy/SEM (HITACHI TM3000, Japan) was used. The coated tomato samples were cut into pieces measuring 2×1 cm. The prepared samples were then mounted onto the SEM sample holder. Once secured, the samples were adjusted and analyzed using SEM to investigate the surface structure and morphology of the applied coating [14].

Image processing of tomato

The image processing technique included 3 main stages: image acquisition, image segmentation, and feature extraction, conducted using MATLAB software to ensure precision and reproducibility. Camera with a 12 MP resolution and aperture $f/1.8$ was used to capture images of the tomatoes. Tomatoes images were captured under controlled lighting conditions to minimize shadows and reflections. A neutral white background was used to enhance contrast, and the camera was positioned at a fixed distance from the object to ensure consistency (± 20 cm). The captured images were then saved in an appropriate format (i.e. JPEG) for further processing. Image segmentation was conducted to separate the tomato image from the background using color-based clustering approach in the CIE-LAB color space for enhanced accuracy. In the stage of feature extraction, key color features were extracted from the segmented tomato region, including: RGB Values, HSV Values, CIE-LAB Values, and Gray-Level Values.

Data analysis

The software package of SPSS version 22.0 (SPSS Inc. USA) was used to perform statistical analyses. A 2-way ANOVA with a significant difference at a 5 % level of probability was performed. If there was a significant difference in the results, the analysis was continued to the Duncan multiple range test. Results of the analysis were expressed as mean \pm standard deviation ($X \pm SD$).

Results and discussion

The application of edible coating contributes to preserving the quality and extending the shelf life of fruits because it prevents the interaction between the fruits and the environment. The coating acts as a barrier for minimizing of loss of moisture and reducing the oxygen availability. In order to perform as expected, the coating should be water-resistant, uniformly covers the fruits, reduce water vapor permeability, keep the structural integrity, and improve the fruits' appearance and non-sticky [1]. Characteristics of the edible films produced from the treatments are presented in the **Figure 1**.

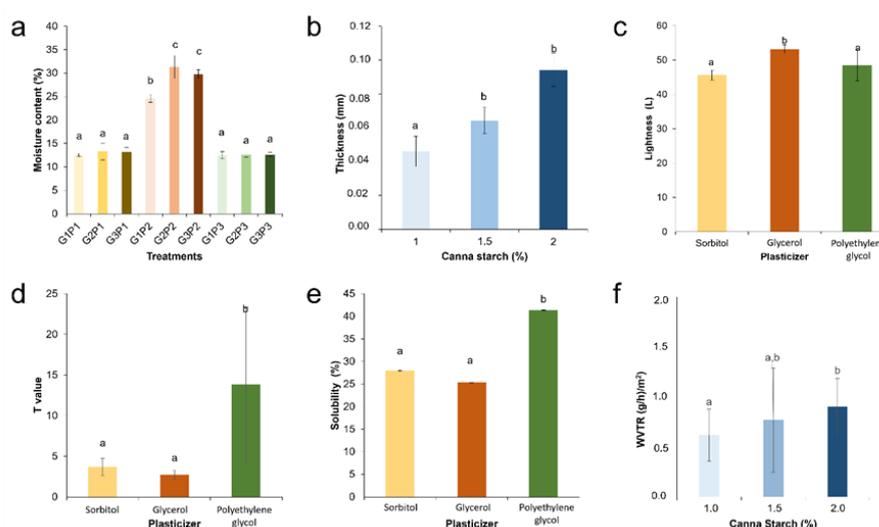


Figure 1 Physical and chemical characteristics of canna starch-based edible films. (a) Moisture content of edible films with different starch concentration (G1 = 1.0 %, G2 = 1.5 %, G3 = 2.0 %) and types of plasticizers (P1 = sorbitol, P2 = glycerol, P3 = polyethylene glycol). (b) Thickness of edible film for each canna starch concentration with addition of 30 % sorbitol. (c) Lightness, (d) optical properties, and (e) water solubility of edible film made of chosen starch concentration with different types of plasticizers. (f). Results are expressed as mean \pm standard deviation. Different superscript letters indicate a significant difference ($p < 0.05$) among different treatments.

Moisture content

An edible film with low moisture content is preferred because it enhances the shelf life of coated products. Canna starch edible films plasticized with glycerol exhibited significantly higher moisture content compared than those plasticized with sorbitol and polyethylene glycol (**Figure 1(a)**). This finding aligns with the previous research on potato starch-based biodegradable films using sorbitol and glycerol as plasticizers [35]. The increased moisture content in glycerol-plasticized films can be attributed to glycerol's ability to form hydrogen bonds, acting as a water-retaining agent [31]. Glycerol's hygroscopic nature

further explains its moisture retention properties [19]. Besides that, moisture content may slightly increase due to hydrogen bonding between water molecules and hydroxyl groups in the biopolymer [40]. Higher glycerol concentrations in the film-forming solution intensify glycerol-water and glycerol-polymer bonds interactions, leading to greater moisture absorption, increased thickness, and structural changes in the biopolymer [40,41].

Thickness

Edible films containing 1.5 or 2 % of canna starch were significantly thicker than those containing 1.0 %

starch (**Figure 1(b)**). However, the interaction between canna starch concentration and plasticizer types did not result in significant differences in thickness among treatments. Film thickness is primarily influenced by the composition and concentration of materials. An increase in the concentration led to thicker films due to the higher concentration of dissolved solids in the film-forming solution. This result is corroborated by the prior reports by Zhang *et al.* [41] and Fransiska *et al.* [42], who reported that higher material concentration yielded thicker edible film made from gum gathi and organic powdered cottonii (OPC). For biodegradable film used in food packaging, adequate thickness is crucial to provide effective protection for food [26]. Conversely, film intended for coating applications should be relatively to ensure uniform adherence to the fruit's surface.

Lightness

A higher value of lightness (L^*) is desirable for films applied as a coating on fruit because it helps maintain the fruit's nature appearance, ensuring the coated fruit closely resembles uncoated ones [14]. It is important because appearance often forms the consumer's 1st impression. Lightness, on the other hand, can contribute to glossiness, resulting in vibrant colors and an improved visual appeal [43], which is likely to attract more consumer attention in displays.

Films plasticized with glycerol exhibited higher L^* value compared to those with sorbitol and polyethylene glycol (**Figure 1(c)**). Meanwhile, there were no significant differences found as affected by the interaction between canna starch concentration and the plasticizer types. Previous studies reported that both glycerol and sorbitol had a slight effect on film lightness [44].

In the present research, the L^* value was found to be less than 60, significantly lower than the 89 - 91 range reported for gum ghatti films [41]. It could be due to the inherent characteristics of the canna starch, tends to exhibit an opaqueness after heating [22]. Moreover, the presence of polyphenols in canna starch, which may oxidize during processing, contribute to the darker appearance of the films [7]. In addition, unmodified canna starch from various varieties is prone to a weaker retrogradation process [45], potentially resulting in lower lightness values [41].

Opacity

The film's transparency is determined by the reflectance caused by light scattering within its structure. A film with a low transparency is considered to have a high opaqueness level [19]. A transparent film is generally preferred for coating fruits as they enhance the product's appearance. On contrary, opaque films are required for packaging lipid-rich foods, which are prone to oxidative degradation catalyzed by UV radiation, such films effectively block UV radiation [19]. In the present research, the solution was designed for fruit coating applications, which require films with low opacity.

The opacity of starch-based films arises from the gelatinization process, which produces an opaque gel [26]. A high value of T indicates a lower degree of film transparency. Figure 1d shows that films incorporating sorbitol and glycerol as plasticizers demonstrated superior transparency compared to those with polyethylene glycol [35]. However, the interaction between canna starch concentration and plasticizer type did not result in significant differences in film transparency. A previous study comparing glycerol and sorbitol as plasticizers also found no significant effect of these plasticizers on films' transparency [39]. Additionally, higher amount of starch likely contributed to increased opacity due to enhanced gelatinization [26].

Water solubility

Water solubility is an important characteristic of edible film, reflecting their ability to dissolve in water and their degree of hydrophilicity. For the coating of fruits, low water solubility is required because it serves as a barrier to moisture movement from the fruits to the surrounding atmosphere [26]. **Figure 1(e)** displays that films plasticized with polyethylene glycol exhibited higher solubility compared to other treatments. Meanwhile, a previous report presented that there is no significant effect of different plasticizers (sorbitol, glycerol, and polyethylene glycol) on the solubility of films produced from the organic powdered cottonii (OPC) [42]. For films made from the gum ghatti, however, the addition of sorbitol resulted in higher solubility than glycerol, with increased plasticizer concentrations further enhancing film solubility.

Film solubility is largely influenced by the amount and type of plasticizer used. Plasticizers disrupt

interactions between biopolymer molecules and act as hydrophilic agents, increasing the film's affinity for water. This increased hydrophilicity promotes greater water absorption and dissolution, thereby affecting the film's overall solubility [41].

Water vapor transmission rate (WVTR)

WVTR is a key indicator a film's barrier property, representing the amount of water vapor that permeates through a unit area of the material over a certain time [26]. As shown in Figure 1f, films produced with 1 % canna starch exhibited a lower WVTR compared to other formulations. However, no significant differences in WVTR were observed due to the interaction between canna starch concentration and the different types of plasticizers. The addition of hygroscopic materials, such as starch, increases the film's affinity for water molecules [35]. This, in turn, enhances water diffusion, and results in higher water vapor permeability [35]. In addition, decreasing the WVTR of films made from canna starch can be achieved by incorporating protein extract [23]. This extract, rich in myofibrillar protein, contains fibrous molecular chains that help form a more compact and solid film matrix structure, thereby improving the barrier properties.

Selected treatment

Edible coatings are applied to the surface of fruits to maintain the quality and extend shelf life. To be effective, these coatings must exhibit low moisture content, sufficient thickness, high lightness and

transparency, low solubility, and controlled WVTR. Based on the evaluation of these parameters, the selected treatment was G1P1, a coating produced from 1 % of canna starch with sorbitol as the plasticizer. This film demonstrated key characteristics, including 12.5 % moisture content, 0.05 mm thickness, 24.8 % water solubility, 4.17 (g/h)/m² WVTR and an L* value of 46.

Tomato, a widely cultivated agricultural product, is highly perishable because of its high moisture content and soft peel. Postharvest losses of tomatoes in developing countries can reach up to 30 % [3]. Therefore, the application of edible coating on tomatoes offers a promising solution as a safer, more cost-effective, and environmentally friendly treatment to extend shelf life.

Application of coating on tomato

The coatings were applied to tomatoes using 2 different methods: spraying (with a 1 mm nozzle) and fogging. **Figure 2** presents the lightness, represented by L* value, of the tomatoes' surface. Neither spraying nor fogging significantly improved the appearance of the tomatoes. Previous research has shown that applying coatings to strawberries resulted in a visually brighter and more reddish compared to uncoated ones, with the coating applied using a sterile brush [14]. In the present research, the coating application did not contribute to a brighter surface because of the inherent characteristics of the coating solution, which tends to exhibit an opaque appearance.

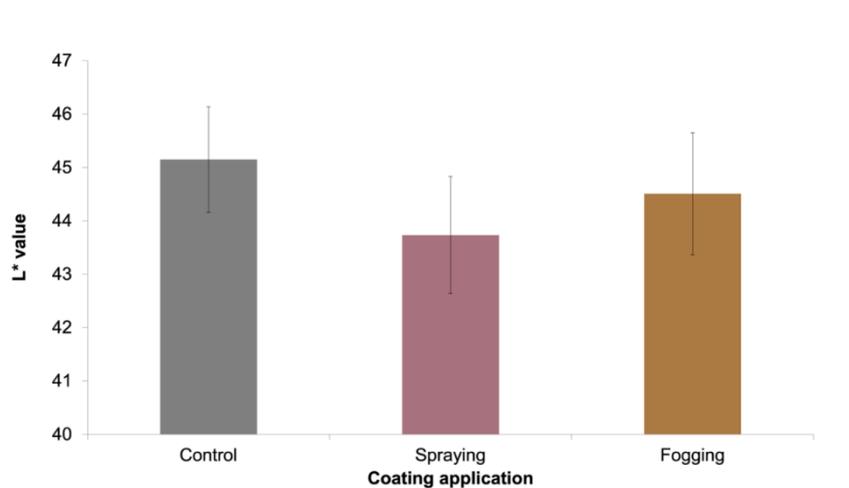


Figure 2 Lightness of tomatoes' surface. Comparison between L* value on control sample with spraying and fogging technique for chosen coating solution (1.0 % canna starch and 30 % sorbitol).

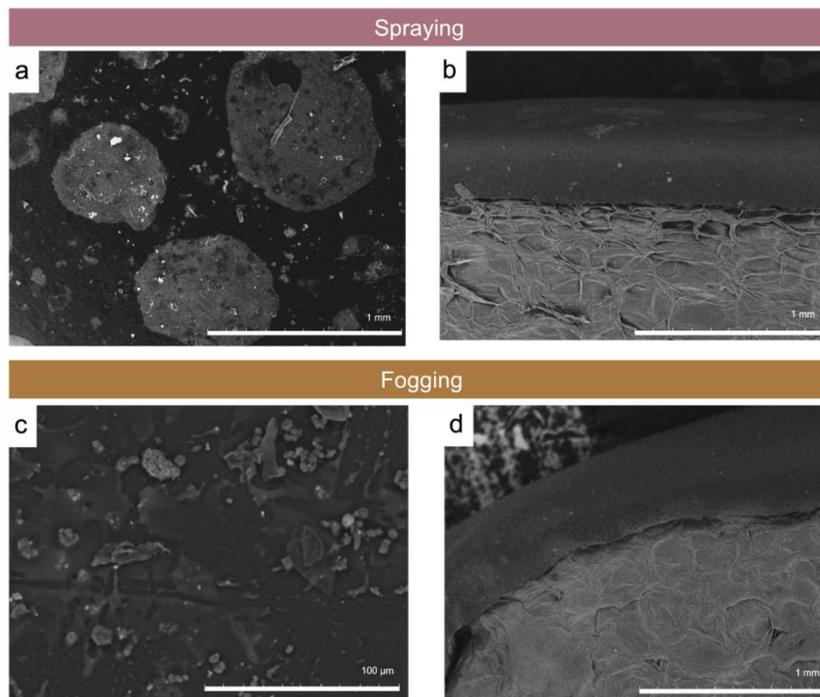


Figure 3 Scanning electron microscopy images of the outer surface and cross-sectional tomato peel. Tomatoes were coated by spraying (a, b) or fogging (c, d) method with coating solution made from chosen formulation above.

The morphology of the coated tomato surface was observed using SEM to analyse the structural details of the coating on the tomato peel. **Figures 3(a)** and **3(c)** show the smoothness and droplet size of the coating. The results revealed that the coating applied *via* the fogging method was more homogenous compared to the spraying method, with smaller droplets that had a more uniform size.

However, the cross-sectional images in **Figures 3(b)** and **3(d)** did not clearly demonstrate the presence of the coating layer on the tomato surface. For future research, the use of staining techniques or the gold-coating method [46], during SEM analysis is recommended to provide a more detailed visualization of the coating on the tomato surface.

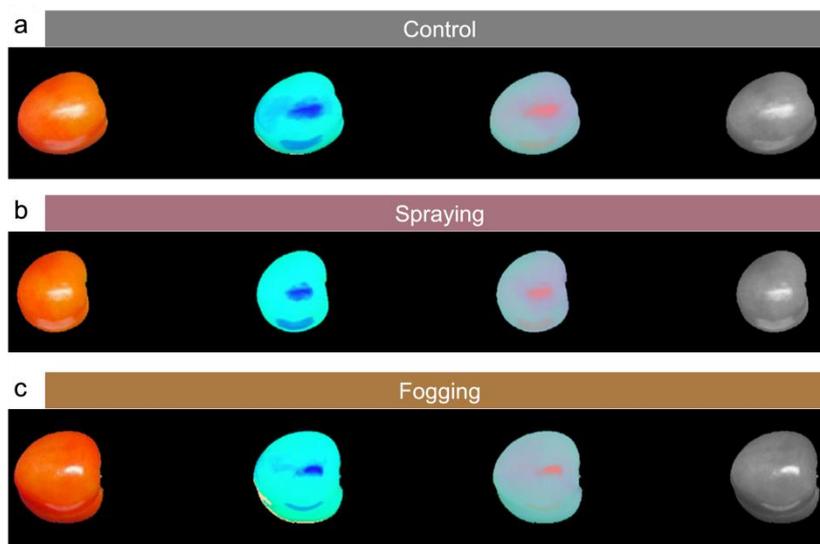


Figure 4 Image analysis of the tomato surface. RGB, HSV, lab object, gray scale images of (a) control sample and coated sample by (b) spraying and (c) fogging methods.

Figure 4 presents the image analysis of the tomatoes coated using spraying (b) and fogging (c) methods compared to the control (a). Each series consists of RGB object images, HSV object images, lab object images, and gray scale images. Image processing has been widely applied for non-destructive evaluation of agricultural product quality. The appearance and color of the peel are crucial sensory factors in determining consumers preference. To provide more accurate assessments of the product, a computer vision system should be used to examine the fruit's appearance at the pixel level [47].

In this research, image processing utilized reflected light that hits the surface of an object and was captured by the camera sensor, then displayed digitally. Digital images containing color information for each pixel were processed using image processing algorithms to achieve the desired results. The use of image analysis to evaluate the quality of fruits with coating application is recommended by previous report [48]. The image analysis of the samples in **Figure 4** did not show significant differences between the uncoated and coated tomatoes. These findings indicate that the canna starch-based coating shows potential for use on tomatoes. To further evaluate its effectiveness, sensory evaluation is recommended to assess consumer acceptance. Moreover, examining the effect of the coating on tomatoes' quality attributes during storage is important for its practical application.

Conclusions

The results showed that starch concentration significantly influenced moisture content, thickness, and water vapor transmission rate, while the type of plasticizer affected lightness, water solubility, and opacity. The interaction of starch concentration and type of plasticizer notably affected moisture content. Based on these findings, the selected treatment was the coating produced with 1 % starch concentration and sorbitol as a plasticizer. The application of this canna starch-based coating using the fogging method resulted in a uniform coating on the tomatoes' surface. Further research is recommended to evaluate the effect of this coating on the quality attributes of tomatoes during storage, providing deeper insights into its potential in extending the shelf-life of tomatoes.

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