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Polysaccharide-based layer-by-layer edible coatings for shelf-life extension of fresh tomatoes

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High yield loss in tomatoes due to suboptimal post-harvest handling and storage demands innovation in preservation technology that is effective and environmentally friendly. Here, we develop polysaccharide-based layer-by-layer edible coatings made of chitosan and hydroxypropyl methylcellulose (chitosan/HPMC) and evaluate their effectiveness for extending the shelf-life of fresh tomatoes using three different application methods (*i.e.*, dipping, air-gun spraying, and manual air-pressure spraying). The coating films are formulated and characterized based on physical, mechanical, and permeability properties. The coated tomatoes have a significantly improved storability of 12 days longer than their uncoated counterpart. Among the investigated application methods, air-gun spraying proves to be the most effective, consistent, and practical method for implementing chitosan/HPMC layer-by-layer on fresh tomatoes. It can limit final weight loss to 7.27%, maintain firmness at 5.44 MPa, and retain ~68% of the total soluble solids after 24 days of storage. These findings provide an important foundation to design efficient edible coating strategies for sustainable post-harvest preservation.

Sustainability spotlight

This study contributes to sustainable food technology by advancing eco-friendly post-harvest preservation methods. The polysaccharide-based edible coatings developed here are biodegradable, non-toxic, and derived from renewable resources, offering an alternative to synthetic packaging. By effectively extending the shelf life of fresh tomatoes while reducing reliance on plastic-based storage solutions, this approach supports waste reduction, resource efficiency, and improved food system resilience.

1 Introduction

Improper handling, inadequate storage, and post-harvest limitations in tomato farming lead to yield reductions of 20% to more than 40%.[1] This loss not only reduces food availability but also disrupts market stocks and economic stability and causes a waste of resources and an increase in carbon footprint.[2–4] Therefore, innovations are needed to overcome these problems, especially for the development of post-harvest preservation technology that is able to extend the storage period and reduce fruit spoilage.[5–10] One solution that has been offered and developed by many researchers is the utilization of edible coatings because they can be an ideal, environmentally friendly alternative to reduce the use of conventional plastics.[11–13]

Edible coating is a widely used strategy to protect perishable fresh horticultural products,[14–17] such as citrus,[18–20] mango,[21] pomegranate,[22] and mushroom.[23] The utilization of edible coatings can prevent weight loss, delay physiological aging, and maintain product quality during storage.[15,16] Edible coatings applied to the surface of the fruit can regulate respiration and transpiration and act as a barrier to pathogenic microorganisms.[24–27] During their development, edible coatings have been made using natural biopolymers, including proteins, lipids, and polysaccharides, because of their abundance in nature and good biocompatibility.[6,9,28–35] Among the available polysaccharides, hydroxypropyl methylcellulose (HPMC) is one of the most widely used due to its ability to form thin films and repel moisture.[36–38] Unfortunately, the use of a single biopolymer is often found to result in low coating performance. Thus, an edible coating composed of more than one material

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has emerged as a promising solution to unlock the potential of the coating film having excellent gas permeability control, mechanical strength, and antimicrobial properties.[39]

From previous studies, a combined coating film comprising chitosan and HPMC (chitosan/HPMC) has shown synergistic performance, where chitosan acts as an antimicrobial agent[10,40–43] and HPMC serves as a film-forming matrix.[44,45] To realize this film, the layer-by-layer deposition method is considered promising because it can deploy oppositely charged biopolymers sequentially to form uniform layers. Although chitosan/HPMC layer-by-layer edible coating offers various benefits (*e.g.*, chitosan not only improves the mechanical resistance of the film structure but also regulates the gradual release of active compounds),[22,40,42,46–48] it still faces various challenges, such as optimization of mechanical strength, applicability, and production on an industrial scale.[45,49,50] Previous studies have used dipping as a typical method to apply edible coatings to fruits.[45,46,51–55] Despite its simple process, the dipping method still has several limitations (*e.g.*, low solution efficiency, inhomogeneous coating distribution, possible cross-contamination, poor scalability, and low reproducibility).

Here, we introduce a comparative evaluation of new spraying-based techniques (*e.g.*, air-gun spraying and manual air-pressure spraying) alongside conventional dipping to demonstrate how application methods influence coating performance, film homogeneity, and shelf-life extension on fresh tomatoes. Furthermore, by monitoring various properties of the coated tomatoes during long-term storage (*i.e.*, weight loss, firmness, total soluble solids, vitamin C, and color), a more comprehensive understanding of the effects of different coating application methods on the fruits can be obtained. As a result, this study can be used as a basis for designing efficient coating strategies for sustainable post-harvest preservation.

2 Materials and methods

2.1 Materials

For the main materials, hydroxypropyl methylcellulose (HPMC) powder (purity ≥99%, viscosity grade ~3000 mPa S) and chitosan flakes were purchased from Prima Chemical Store (Purwokerto, Indonesia) and Chitafood (Yogyakarta, Indonesia), respectively. Since the molecular weight and degree of deacetylation were not specified in the product documentation, we used viscosity grade (for HPMC) and commercial grade (for chitosan) as the primary characterization parameters in this study. Glacial acetic acid purchased from Prima Chemical & Packaging was used as chitosan solvent. Tomato (*Solanum lycopersicum* 'Servo') samples were hand-picked at uniform maturity (light red to red) and with consistent dimensions (diameter of 4–5 cm and weight of 40–70 g) from local farmers in Sumbang, Banyumas, Central Java, Indonesia. Sodium chloride (NaCl) from Merck, Darmstadt, Germany, was used to control the environment in the permeability test. For vitamin C assessment, two reagents—amylum (Merck, Germany) and iodine (Sigma-Aldrich, Singapore)—were employed. All materials and samples were used as received without any further purification or pre-treatment.

2.2 Edible film production

HPMC powder was dissolved in distilled water that had been heated at 45–50 °C for ~10–15 minutes with three different concentrations of 0.4%, 0.8%, and 1.2% (w/v). After adding HPMC, the mixed solutions were stirred using a hot plate stirrer for ~15 minutes until 60–70 °C and then cooled to 30 °C to produce the HPMC solution. Separately, chitosan solutions were prepared by dissolving chitosan powder with three different concentrations of 0.5%, 1.0%, and 1.5% (w/v) in 1% (v/v) acetic acid. The chitosan solutions were then stirred using a hot plate stirrer (T = ~50 °C for ~15–20 minutes) and subsequently cooled to 30 °C until the solutions became clear. Both HPMC and chitosan solutions were then poured into separate plastic molds with a thickness of ~2–3 cm and then dried in a food dehydrator at ~30 °C for 12–14 hours to obtain HPMC and chitosan single-layer films. In addition to those single-layer films, bi-layer chitosan-combined HPMC (chitosan/HPMC) films were prepared using a layer-by-layer technique by depositing the chitosan and HPMC solutions sequentially. All fabricated films are shown in Table 1 and illustrated in Fig. 1a. All concentrations used in this study were determined from preliminary trials based on previous studies.[45,56] These concentrations were selected to achieve proper polymer dispersion and coating uniformity without causing excessive viscosity that may hinder spraying performance.

2.3 Edible film characterization

Each single-layer film was placed on a white paper surface for color analysis based on the ASTM E1347 standard using a color reader (Chnspec CS-10) at 10 different points. The results were represented as L, a^*, and b^* values and subtracted from the white plate standard to obtain ΔL, Δa, and Δb, which were then calculated to obtain the ΔE value using eqn (1):

$$\Delta E = \sqrt{\Delta L^2 + \Delta a^2 + \Delta b^2} \quad (1)$$

After being cut to a size of 4 cm × 1 cm, the film was measured in terms of its thickness (d) and subsequently inserted into a UV-Vis spectrophotometer to read the absorbance at a wavelength of 600 nm (A_{600}) for obtaining its opacity using eqn (2):[48]

$$\text{Opacity}(\%) = \frac{A_{600}}{d(\text{mm})} \quad (2)$$

Water vapor permeation (WVP) values were generated using the ASTM E96 standard with a circular sample with a diameter

Table 1 Fabricated films based on hydroxypropyl methylcellulose (HPMC) and chitosan having different layer types

Material base	Layer type
HPMC	Single-layer
Chitosan	Single-layer
Chitosan/HPMC	Bi-layer

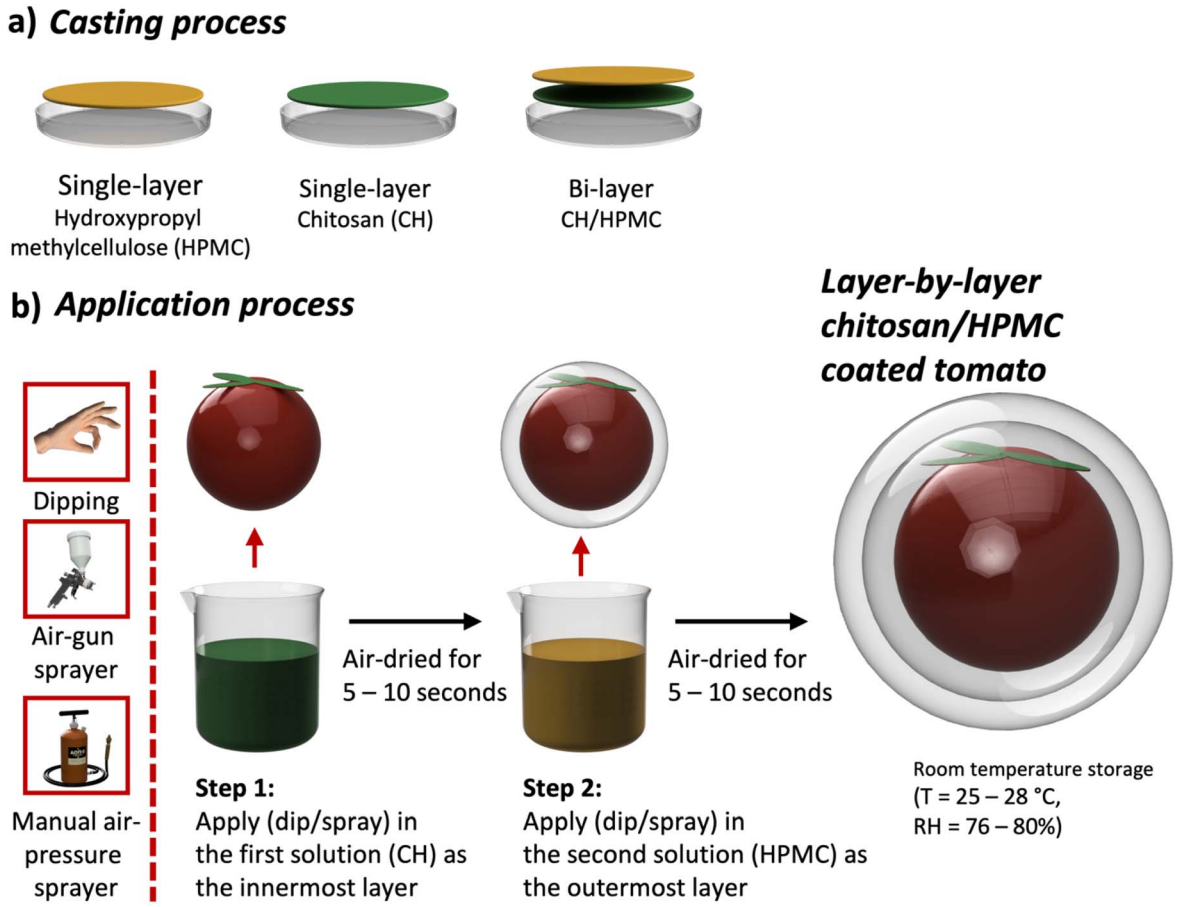

Fig. 1 Production of edible chitosan/hydroxypropyl methylcellulose (HPMC) films and coating applications. (a) Casting process to fabricate single-layer hydroxypropyl methylcellulose (HPMC), single-layer chitosan, and bi-layer chitosan/HPMC films. (b) Three different application processes (dipping, air-gun spraying, and manual air-pressure spraying) of single-layer hydroxypropyl methylcellulose (HPMC), single-layer chitosan, and bi-layer chitosan/HPMC on a tomato.

of 4.5 cm. A film sheet was used for the cup containing silica gel and placed in a modified chamber using saturated sodium chloride (RH = 60–70%, $\Delta P = 4245$ Pa). The cup was weighed (Δw) regularly at certain intervals (Δt) and calculated using eqn (3):

$$\mathrm{WVP}\left(\mathrm{g\ s^{-1}\ m^{-1}\ Pa^{-1}}\right) = \frac{\Delta w(\mathrm{g}) \times d(\mathrm{m})}{\Delta t(\mathrm{s}) \times A(\mathrm{m^2}) \times \Delta P(\mathrm{Pa})} \quad (3)$$

To evaluate the mechanical properties of the film, tensile strength (TS) and elongation at break (EAB) values were determined using ASTM D882 standards. The mechanical strength testing was carried out using a rectangular membrane cut to dimensions of 10 cm × 2 cm and performed on a texture analyzer (Stable Microsystems TA-XT Plus) under conditions of 50 mm initial grip separation (L), 50 mPa force, and 50 mm $\mathrm{s^{-1}}$ constant velocity. Force (F) and final length before break (L') data were recorded as test result data. The TS and EAB values were then calculated using eqn (4) and (5), respectively:

$$\mathrm{TS(MPa)} = \frac{F(\mathrm{N})}{d(\mathrm{m}) \times L(\mathrm{m})} \quad (4)$$

$$\mathrm{EAB}(\%) = \frac{\left(L - L'\right)}{L} \times 100\% \quad (5)$$

Surface morphology, chemical composition, and wettability were studied on the film to obtain its optimum coating order. The surface morphology of the edible coating film was observed using scanning electron microscopy (SEM, JEOL JSM 6510) by cutting the sample into a 2 cm × 2 cm size. Morphological observations were made in the form of surface structure and uniformity. The chemical composition of the film was analyzed using Fourier-transform infrared (FTIR) spectroscopy (Thermo Scientific Nicolet iS10) in the range of 400–4000 $\mathrm{cm^{-1}}$. The obtained spectra were used to identify the functional groups of the films and observe the molecular interactions between HPMC and chitosan, as well as evaluate the coating integrity. The wettability of the film was evaluated in terms of its water contact angle (WCA) using the sessile drop method (Fumalife). The 6 μL of deionized water was gently placed on the film surface using a micro-syringe, and images were captured at 0–30 s intervals using an optical camera. The resulting images from the WCA observation were then analyzed using ImageJ software. The

WCA was measured in three repetitions ($n = 3$) for each sample, and the standard deviation (SD) of the repetition calculation was used as the measurement error.

2.4 Tomato applications

Tomatoes were harvested from local farmers (Sumbang, Banyumas, Central Java, Indonesia) and strictly selected based on color (light red to red), appearance (no physical damage), a weight of 40–70 g, and a diameter of 4–5 cm. To ensure that microbial effects did not interfere with the evaluation of coating performance, all tomatoes were carefully pre-treated by washing and sanitizing prior to coating application, following standard post-harvest handling practices.[57] The selected tomatoes were then divided into nine groups. The different groups represented treatment differences in terms of the sequence (*i.e.*, single-layer HPMC, single-layer chitosan, and bi-layer chitosan/HPMC) and application technique (*i.e.*, dipping, air-gun spray, and manual air-pressure spray). Tomatoes were washed and air-dried prior to coating, as illustrated in Fig. 1b. All tomato samples were then put into plastic trays and covered with plastic nets to prevent pest infestation after receiving the treatments. Those samples were stored at room temperature ($T = 25$–28 °C; RH = 76–80%; with no light exposure). Observations were carried out for 24 days, and evaluations were made every 6 days.

2.5 Tomato assessment

Tomatoes were evaluated based on physical (*i.e.*, color, weight loss, and firmness) and chemical (*i.e.*, total soluble solids and vitamin C) properties every 6 days. First, the surface color was observed using a color reader (Chnspec CS-10). Observations were made at 5 random places on the surface of the tomato. The data obtained in the form of L, a^*, and b^* were calculated as Minolta color values using eqn (6):

$$\text{Minolta color } (\Delta E) = \frac{a^*}{b^*} \tag{6}$$

These results were then compared with the United States Department of Agriculture (USDA) classification of tomato fruit maturity stages in Table 2. Tomatoes were weighed using an analytical balance to determine the final weight on the day of observation (b) and compared with the initial weight from the first day of storage (a). Based on these values, the amount of weight loss (WL) can be determined based on the calculation using eqn (7):

$$\text{WL}(\%) = \frac{a(\text{g}) - b(\text{g})}{a(\text{g})} \times 100\% \tag{7}$$

To determine the firmness of the tomatoes, a texture analyzer (Stable microsystem TA.XT plus) with a 5 mm diameter cylindrical probe was used with defined settings (*i.e.*, pre-test speed of 3.0 mm s^{-1}, test speed of 1.0 mm s^{-1}, post-test speed of 30 mm s^{-1}, and penetration distance of 3 mm). For chemical analysis, tomatoes were cut and crushed into pulp. A total of 1–2 mL of tomato pulp was then added to a digital hand refractometer (Alla France 950.0100 PPT-ATC) to obtain the total soluble solids (TSS). TSS data were collected in units of °Brix. To determine the vitamin C value, tomato pulp was diluted in distilled water (1 : 10 v/v) and filtered using filter paper to produce tomato extract. A total of 20–30 mL of tomato extract was mixed with 1% amylum indicator and titrated using iodine 0.01 N until the color turned blue. The volume of iodine (V_{iodine}) was used to calculate vitamin C using eqn (8):[45]

$$\text{Vitamin C } (\%) = V_{\text{iodine}} \times 0.88 \times \text{dilution ratio} \tag{8}$$

2.6. Statistical analysis

Data obtained from observation were analyzed using one-way analysis of variance (ANOVA) with a $p \leq 0.05$ significance level and a *post hoc* test using Duncan's multiple range test (DMRT) for parametric data. Non-parametric data were analyzed using the Kruskal–Wallis test with a significance level of $p \leq 0.05$ and the Mann–Whitney U-test as a *post hoc* test method. All statistical analyses were conducted using Statistical Product and Service Solutions (SPSS) software.

Table 2 United States Department of Agriculture (USDA) classification of tomato fruit maturity stages. Minolta color values are correlated to tomato color stages

Minolta color values (a^*/b^*)	USDA tomato color stages
−0.59 to −0.47	Green
−0.47 to −0.27	Breaker
−0.27 to 0.08	Turning
0.08 to 0.60	Pink
0.60 to 0.95	Light red
0.96 to 1.21	Red

3 Results and discussion

3.1 Edible film characteristics

Fig. 2a shows the water vapor permeability (WVP) test results of HPMC films with various concentrations of 0.4%, 0.8%, and 1.2%. The WVP values obtained are 6.98×10^{-11}, 5.80×10^{-11}, and 3.85×10^{-11} g s^{-1} m^{-1} Pa^{-1} for HPMC concentrations of 0.4%, 0.8%, and 1.2%, respectively. The WVP value of the HPMC films decreased as the concentration increased. This phenomenon may occur when the concentration of the matrix solution is increased and the film structure becomes more compact, limiting its interaction with water.[58] The water vapor permeability (WVP) test is known as a parameter to observe material and water interaction.[43,59] On the other hand, the oxygen transmission rate (OTR) is typically used as one of the important parameters for evaluating the gas barrier functionality of edible films.[60–63] However, in this study, we focused on WVP as the primary barrier property, since moisture loss is the dominant factor influencing tomato deterioration. Additionally, the polysaccharide coatings applied here are inherently more effective against water vapor transfer compared to oxygen diffusion. Fig. 2b and c show the color and opacity

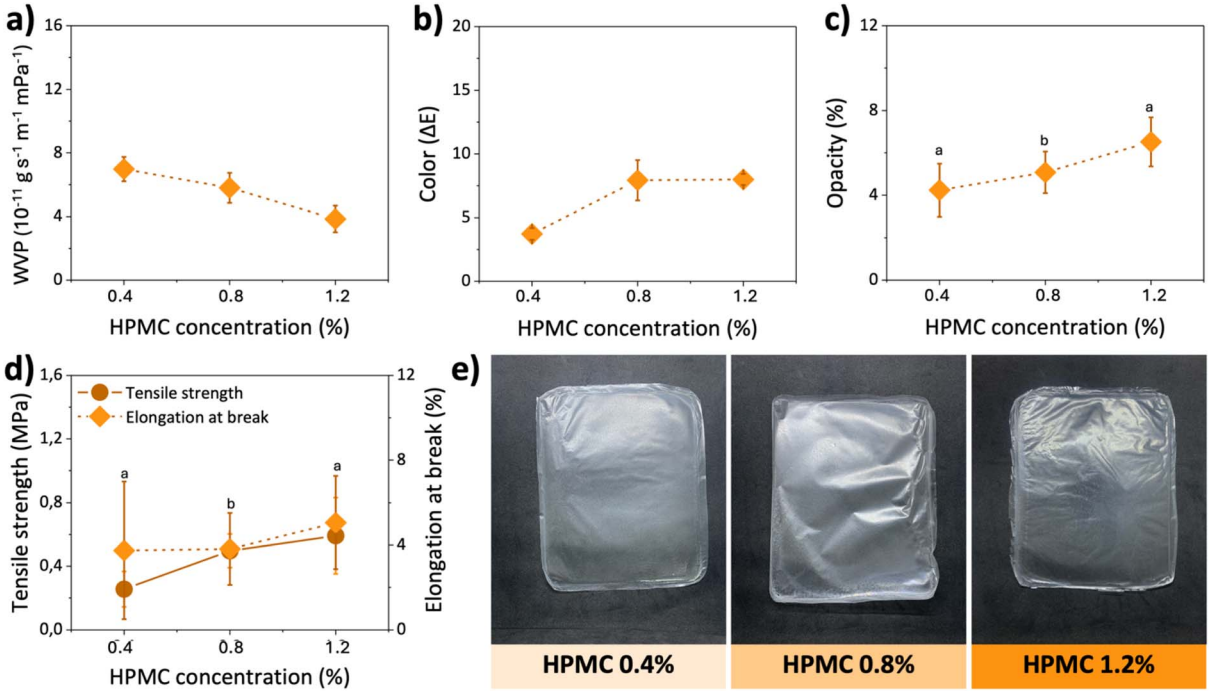

Fig. 2 Hydroxypropyl methylcellulose (HPMC)-based edible film characteristics with various concentrations. (a) Water vapor permeability (WVP) analysis, (b) color difference (ΔE) analysis, (c) opacity measured using 600 nm wavelength, (d) tensile strength and elongation at break, and (e) visual appearance of the fabricated HPMC-based edible film with concentrations of 0.4%, 0.8%, and 1.2%, respectively. Different superscript letters indicate significant differences ($p \leq 0.05$) according to the *post hoc* *t*-test with Duncan's multiple range test (DMRT).

measurement results of the HPMC films. These two parameters (*i.e.*, color and opacity) are crucial to evaluate, as they are influential in the product appearance and good consumer acceptance. As the concentration of HPMC increases, the resulting ΔE value becomes higher. The increase in the ΔE value indicates a decrease in the clarity of the film. These results are then supported by the opacity value, which continues to increase with values of 4.24%, 5.08%, and 6.52% for HPMC concentrations of 0.4%, 0.8%, and 1.2%, respectively. These results show that both parameters increase with increasing concentration of HPMC used in film fabrication, which is believed to be due to an increase in the solid content of the HPMC film.[64]

In addition to the appearance of the edible film, mechanical strength is also an important parameter to be considered, as it affects the film resistance to physical contact in its application as an edible coating. The mechanical strength of the HPMC-based edible film was evaluated based on the tensile strength and elongation at break values, as shown in Fig. 2d. The tensile strength values are (0.26 ± 0.11) MPa, (0.50 ± 0.11) MPa, and (0.59 ± 0.24) MPa, while the elongation at break values are (3.74 ± 3.25)%, (3.82 ± 1.70)%, and (5.06 ± 2.20)%, for HPMC films with concentrations of 0.4%, 0.8%, and 1.2%, respectively. The increase in both values indicates that the mechanical strength of the film rises as the concentration of HPMC used increases. This phenomenon is very likely to occur because, with an increase in the concentration used, the density of the resulting film increases, making the resulting film stronger. The appearance of the fabricated HPMC-based edible film for each concentration can be observed based on the results of the photographs shown in Fig. 2e. The detailed data of HPMC film characteristics can be found in Table S1 in the SI.

Fig. 3a shows the WVP measurement results of chitosan-based edible films with various concentrations, with values of 1.26×10^{-11}, 1.22×10^{-11}, and 1.06×10^{-11} g s^{-1} m^{-1} Pa^{-1} for chitosan concentrations of 0.5%, 1.0%, and 1.5%, respectively. Similar to the WVP results of HPMC-based films, the WVP values of chitosan-based films decreased with increasing concentration, which is believed to be due to the same reason as that of HPMC-based films.[58] However, the WVP value of chitosan-based films for each concentration is much lower than that of HPMC-based films, indicating that the chitosan film has a tighter structure. This phenomenon is supported by chitosan's ability to create a vapor barrier by forming a zigzag path to resist water diffusion.[65] Fig. 3b and c show the color and opacity measurement results of chitosan-based films. Both values are almost equal to those of HPMC-based film and increase with increasing concentration, as seen in HPMC-based films. In contrast to HPMC-based films, the different color pigments of chitosan are believed to be the main factor determining the color and opacity of the fabricated films.[66] Chitosan contains the crustacean-derived pigment astaxanthin. This pigment causes an increase in the ΔE value of the fabricated film, which then results in a decrease in the opacity value when compared to that of the HPMC-based film.

The tensile strength and elongation at break values of the chitosan-based film can be seen in Fig. 3d. The tensile strength values of the chitosan-based film are (0.23 ± 0.07) MPa, $(11.20 \pm$

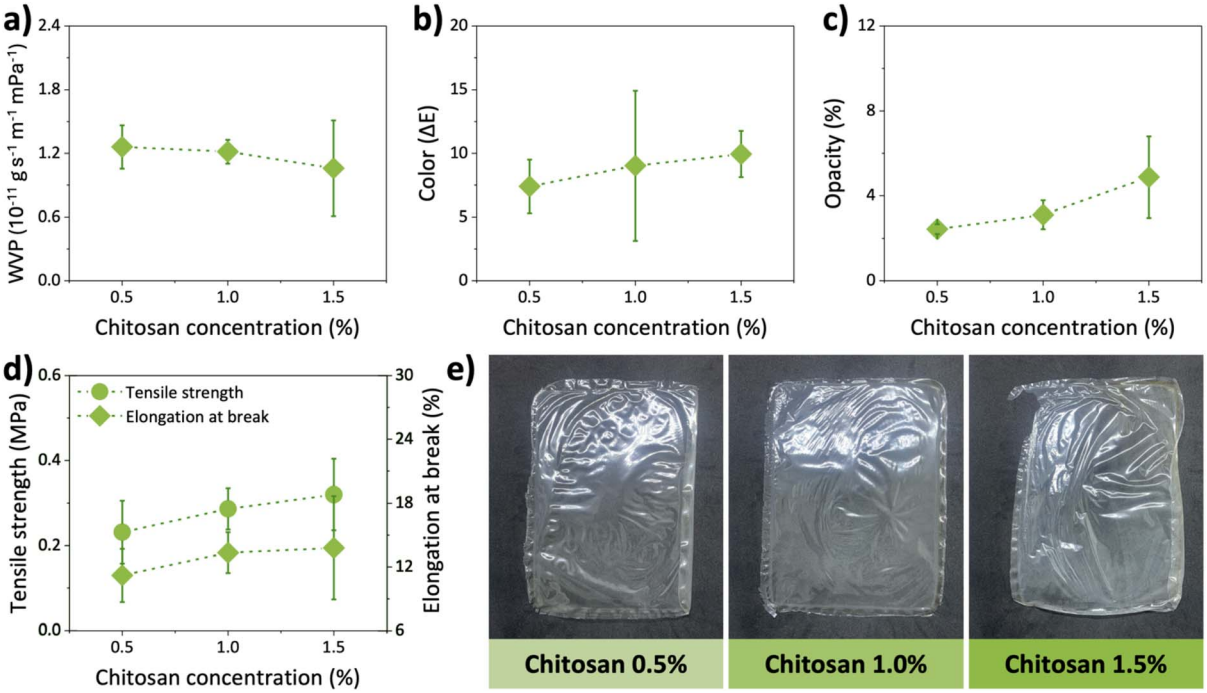

Fig. 3 Chitosan-based edible film characteristics with various concentrations. (a) Water vapor permeability (WVP) analysis, (b) color difference (ΔE) analysis, (c) opacity measured using 600 nm wavelength, (d) tensile strength and elongation at break, and (e) visual appearance of the fabricated chitosan-based edible film with concentrations of 0.5%, 1.0%, and 1.5%, respectively. Different superscript letters indicate significant differences ($p \leq 0.05$) according to the *post hoc* *t*-test with Duncan's multiple range test (DMRT).

2.51) MPa, and (0.29 ± 0.05) MPa, with elongation at break values of (13.34 ± 1.92)%, (0.32 ± 0.08)%, and (13.79 ± 4.87)% for concentrations of 0.5%, 1.0%, and 1.2%, respectively. It is clearly seen that the tensile strength values of chitosan-based films with various concentrations are lower than those of HPMC-based films. In contrast, the elongation at break value of chitosan is much higher when compared to that of the HPMC-based film. These results indicate that chitosan has less strength but much higher elasticity. The mechanical properties are necessary to evaluate because in the practical applications, the polymer may face different stresses.[67,68] The appearance of the chitosan-based film can be seen in Fig. 3e. The detailed values of chitosan film characteristics are presented in Table S2 in the SI.

3.2 Layer-by-layer characteristics

Fig. 4 shows the Fourier-transform infrared (FTIR) spectra of the fabricated edible films. The HPMC monolayer has characteristic peaks at 3415 cm^{-1}, 2902 cm^{-1}, and 1644 cm^{-1} associated with O–H, C–H, and C=O stretching vibrations, respectively, which indicate the presence of hydroxyl, methyl, and carbonyl groups. The peaks in the fingerprint region (500–1500 cm^{-1}) are associated with C–H (1454 cm^{-1} and 956 cm^{-1}) and C–O–C (1327 cm^{-1} and 1052 cm^{-1}) vibrational bands, which are recognized as methyl and ether bonds in the matrix. These results are similar to previous studies.[69] For the chitosan monolayer, the peaks at 3167 cm^{-1}, 1605 cm^{-1}, and 1529 cm^{-1} define O–H and N–H stretching, C=O stretching, and N–H bending vibrations associated with hydroxyl, amide, and amine groups. These findings are in agreement with previously reported investigation results.[45] With the combination of HPMC as the outer surface on chitosan, the same functional groups as the monolayer were found in the spectra of the chitosan/HPMC bi-layer film, namely hydroxyl, amine, methyl, and ether.

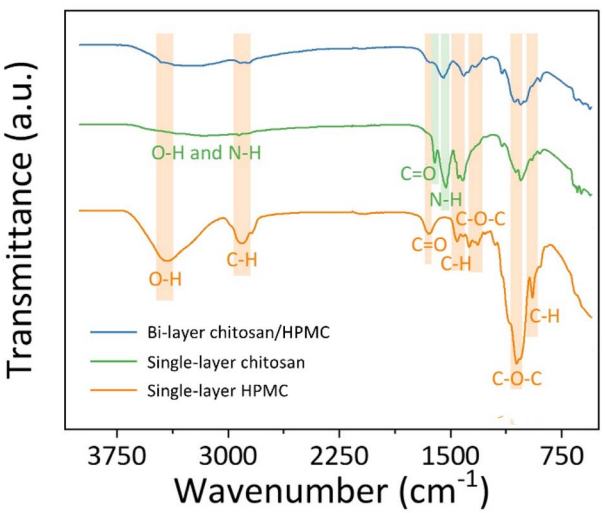

Fig. 4 Fourier transform infrared (FTIR) spectra of various edible films. Three different films were investigated (*i.e.*, single-layer hydroxypropyl methylcellulose (HPMC), single-layer chitosan (CH), and bi-layer chitosan/HPMC).

However, broader peaks near 3000–3500 cm^{-1} were found in the chitosan/HPMC bi-layer, which are responsible for O–H and N–H stretching vibrations. This broadening, accompanied by a slight shift to lower wavenumbers, suggests the strengthening of hydrogen bonding between the hydroxyl group of HPMC and the amino/hydroxyl groups of chitosan. Additionally, noticeable shifts in the amide I (≈1600–1650 cm^{-1}) and amide II (≈1500–1580 cm^{-1}) bands indicate electrostatic interactions between the protonated $-NH_3^+$ groups of chitosan and the hydroxyl-rich HPMC matrix. These spectra confirm that the layer-by-layer assembly enhances intermolecular bonding and contributes to the formation of a more compact and stable bi-layer structure.[65]

The surface morphology of the fabricated films is shown in Fig. 5a according to SEM images. Slight cracks are identified on the single-layer HPMC and rough surfaces are found on the single-layer chitosan and bi-layer chitosan/HPMC. Among them, the HPMC film depicts a more uniform, dense, and smooth surface. Such characteristics can provide a stable and better barrier as a coating layer. While the chitosan film has almost similar results, it is slightly rougher, and some irregularities, such as the presence of small porous structures, are detected. On the other hand, the bi-layer chitosan/HPMC shows a rough and structured surface morphology, potentially indicating a multilayered structure due to electrostatic interactions between layers. In particular, chitosan as the base layer provides a more compact, dense, and less rough foundation, while HPMC as the outer layer results in a smoother and less lumpy surface. Therefore, chitosan/HPMC with HPMC as the outer layer appears to have a better structure. Although morphological properties are not the only crucial factors to determine coating suitability, they still need to be well controlled to accomplish better surface characteristics (*i.e.*, homogeneous, smooth, compact, and uniform). Earlier studies have found that adding plasticizers (*e.g.*, glycerol, sorbitol, and gelatin) in certain amounts can prevent cracks and produce a smoother surface on the films.[57,70] Thus, it can enhance other properties such as better adhesion, less permeation, and separation.

Fig. 5b shows the water contact angle (WCA) test results of all three films (*i.e.*, HPMC, chitosan, and chitosan/HPMC films) to evaluate their wettability. HPMC and chitosan films exhibit different wettability behavior. The HPMC film has a low WCA value of (69 ± 5)°, indicating its hydrophilic properties. This result is consistent with its smooth and continuous surface morphology, which can be one factor that influences surface wettability. On the other hand, chitosan has a higher WCA value of (88 ± 5)°, indicating its more hydrophobic properties. Apart from the rougher surface morphology, the high WCA of the chitosan film is also caused by its chemical structure. Here, chitosan only has amino bonds, making it less likely to interact with water.[71,72]

Meanwhile, the chitosan/HPMC film shows a moderate WCA value (located between those from HPMC and chitosan films), which amounts to (81 ± 5)°. This WCA value indicates that the deposition of chitosan followed by HPMC results in enhanced interfacial interaction, leading to a more uniform film with optimal wettability. Thus, the chitosan/HPMC film could potentially offer a controlled barrier effect, limiting dehydration while allowing gas exchange. These findings suggest that chitosan/HPMC coatings provide optimal conditions between surface wettability and film integrity, making them the most suitable treatment for preserving tomato freshness. Although the mechanistic characterizations (*i.e.*, SEM, FTIR, and wettability) were carried out on standalone bi-layer films rather than tomato surfaces, the results provide a strong indication of the coating behavior when applied to target fruits (tomatoes).

3.3 Application to tomatoes

3.3.1 Weight loss. Fig. 6a shows the weight loss values over 24 days of storage for each tomato coated with the best-performing edible coating (*i.e.*, chitosan/HPMC) using different methods (dipping, air-gun spraying, and manual air-pressure spraying). Unfortunately, the uncoated tomatoes experienced significant weight loss from day 6 and could not be further analyzed on day 12 due to physical deterioration, such as softening, black spots, and excess moisture content. This phenomenon might occur because the uncoated tomatoes were exposed directly to air, which accelerates the transpiration rate.[73] In contrast, the application of an edible coating successfully maintained the integrity of tomatoes until the end of the 24-day observation period.

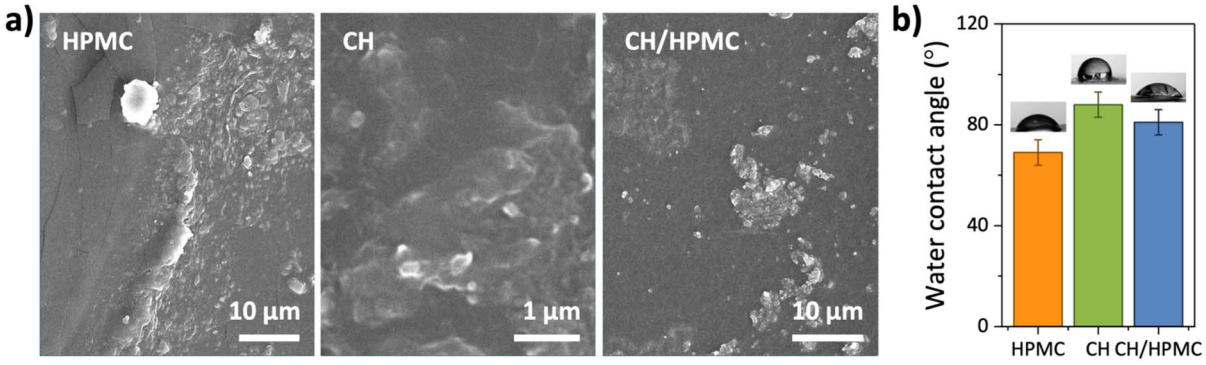

Fig. 5 Morphological characteristics and surface wettability of edible films. (a) Scanning electron microscopy (SEM) images and (b) water contact angle (WCA) of single-layer hydroxypropyl methylcellulose (HPMC), single-layer chitosan (CH), and bi-layer chitosan/HPMC (CH/HPMC).

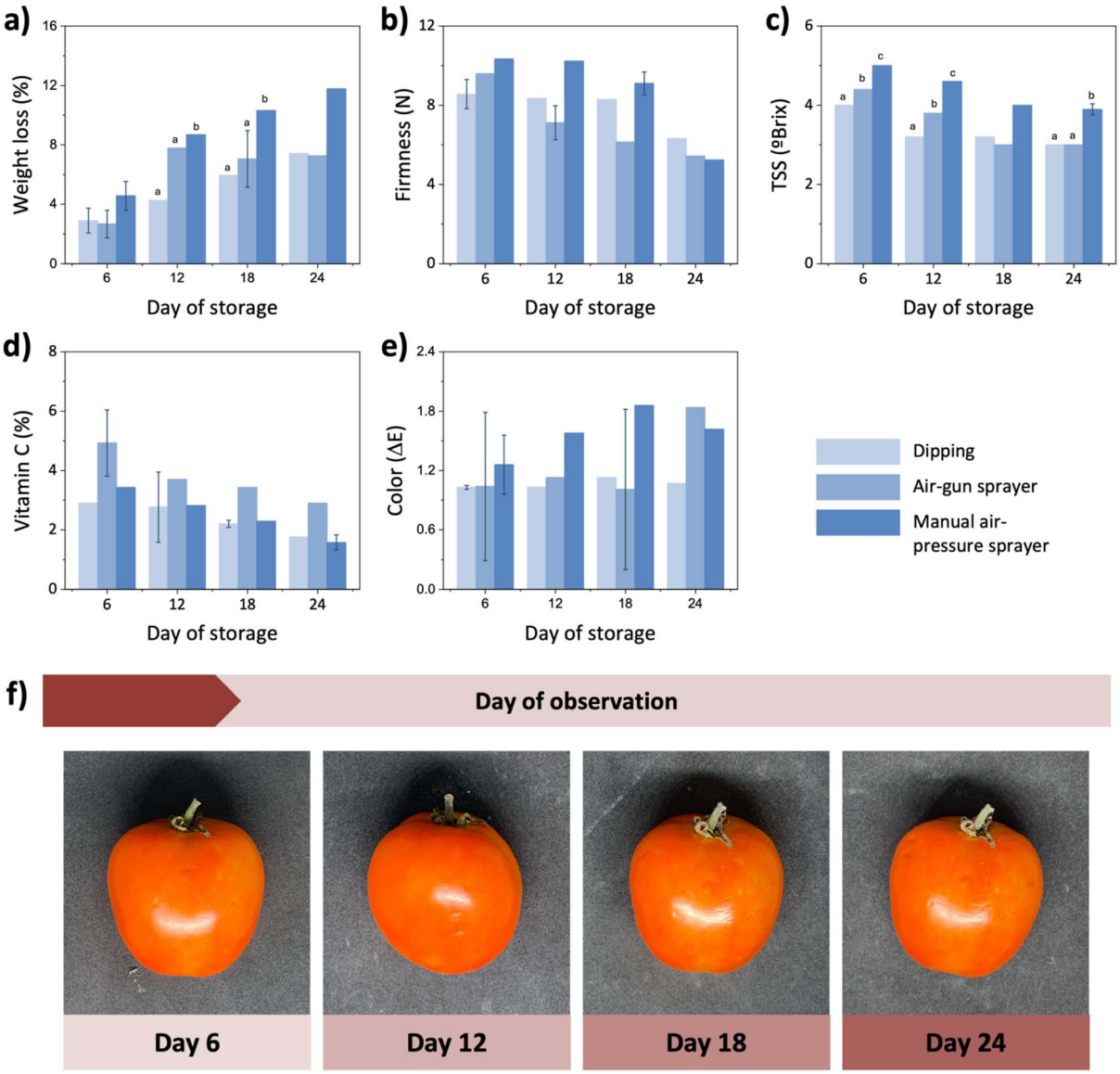

Fig. 6 Physicochemical characteristics of bi-layer chitosan/HPMC-coated tomatoes during 24 days of storage processed by three different coating application methods (dipping, air-gun spraying, and manual air-pressure spraying). (a) Weight loss analysis, (b) firmness results, (c) total soluble solids (TSS) using a refractometer, (d) titratable vitamin C, (e) color analyses using Minolta (a^*/b^*) measurement, and (f) photograph profiles of coated tomatoes showing different color intensities during continuous observation from day 6 to 24. Different superscript letters indicate significant differences ($p \leq 0.05$) according to the *post hoc* t-test with the Mann–Whitney U-test.

The different coating application methods (dipping, air-gun spraying, and manual air-pressure spraying) showed different weight loss patterns. On day 24, the manual spraying method resulted in the highest weight loss of 11.78%, while the dipping and air-gun spraying methods recorded mass losses of 7.42% and 7.27%, respectively ($p > 0.05$). Although the difference was not statistically significant, this phenomenon of different mass losses can be attributed to the variation in the nozzle diameter. Manual air-pressure spraying uses a large diameter nozzle, which tends to produce an uneven and excessive coating, which can accelerate water loss. In contrast, an air-gun with a small nozzle (0.6 mm) produces more homogeneous droplets. This finding is consistent with a previous study,[56] which emphasized the importance of controlling the nozzle size, spraying distance, pressure, and flow rate for optimal coating formation.

3.3.2 Firmness. In general, all tomato samples decreased in firmness over time, whether coated or uncoated. However, the coated tomatoes retained their texture better and lasted up to 12 days longer than the uncoated samples. The decrease in firmness was attributed to the activity of enzymes such as pectin esterase and polygalacturonase, as well as the degradation of the cell wall structure during ripening, resulting in degradation of the cell wall and cellular turgidity.[21,74] Edible coatings act as a barrier against moisture loss and gas exchange, slowing down such enzymatic activities.

The firmness observation in Fig. 6b shows that for each coating method, the firmness value decreases. On day 6, the firmness values were (8.6 ± 0.7) N, (9.6 ± 0.0) N, and (10.3 ± 0.0) N, and on day 24, they became (6.3 ± 0.0) N, (5.4 ± 0.0) N, and (5.2 ± 0.0) N for the dipping, air-gun spraying, and manual air-pressure spraying coating methods, respectively. Although the dipping coating showed the smallest firmness value on day 6 compared to the other methods, it did not decrease significantly on day 24, making it the highest firmness value. Meanwhile, for air-gun spraying, its highest firmness value was found on day 6 and started to decrease significantly on day 24. This phenomenon occurs because the coating film produced by the dipping method possesses a higher thickness than that processed by the air-gun spraying method. Although air-gun spraying can provide a more even coating, it cannot be as thick as the dipping method. This makes tomatoes coated by the dipping method maintain firmness better. The increase in firmness durability observed after layer-by-layer coating is in line with the results of a previous study conducted on Japanese pears (Kosui).[50]

3.3.3 Total soluble solids and vitamin C. Fig. 6c and d display the analysis of total soluble solids (TSS) and vitamin C focused on the performance of chitosan/HPMC coating formulations with three different application methods. Based on the conducted tests, the different application methods have a significant impact on the ability of the coatings to maintain nutritional quality during storage. The manual air-pressure spraying method shows the best performance in maintaining TSS content, with a final value of 3.90 °Brix after 24 days of storage. This value was significantly higher than the dipping and air-gun spraying methods, which each recorded the same TSS of 3.00 °Brix. This indicates that manual air-pressure spraying produces coatings of sufficient thickness to form an effective semi-permeable barrier, without causing excess moisture accumulation or adverse anaerobic conditions.

Meanwhile, the vitamin C content of each method also gives striking results, where the trend shows that the air-gun spraying method has the advantage in maintaining nutrient stability. The trend of the highest vitamin C content from the air-gun spraying method persists until the observation on day 24, which is believed because this method can produce a very thin and even layer that can provide optimal protection to tomatoes, aligning with the weight loss observation. On the other hand, the highest decrease in vitamin C occurs in the dipping method, which is assumed to produce a layer that is too thick and can prevent optimal gas exchange. This excess coating thickness also has the potential to create an internal environment that accelerates vitamin degradation due to oxidative stress or atmospheric imbalance.[57,75]

3.3.4 Color. Fig. 6e shows the color difference (ΔE) of the coated tomatoes according to the Minolta color value. The change in the Minolta color value (a^*/b^*) was used as an indicator of ripeness and visual degradation of the analyzed tomatoes. The highest maturity, corresponding to ready-to-consume tomatoes, was indicated by the "red" color value. Uncoated samples showed a spike in the a^*/b^* value from 0.55 to 1.09 within the first 6 days, indicating rapid ripening due to ethylene production. This can accelerate quality degradation. These samples were not suitable for analysis after day 12.[75] In contrast, the coated samples exhibited a slower color change, which makes the samples viable until the end of the observation day as depicted in Fig. S1 in the SI.

The highest Minolta color value was achieved by the air-gun spraying method (1.84), followed by manual air-pressure spraying (1.62) and dipping (1.07). While all variants are still within the "ripe red" range according to the USDA, the coating produced by the air-gun is more homogeneous and thinner, allowing for reasonable ripening without introducing off-flavor effects or immaturity. The photographs of the coated tomatoes using different application methods are displayed in Fig. 6f. In contrast, thick layers from the dipping end delay ripening due to limited gas exchange and light penetration.[76]

3.3.5 Application method evaluation. Three different methods successfully facilitated layer-by-layer construction of chitosan/HPMC and enhanced the post-harvest and storability of fresh tomatoes, offering a shelf-life extension of 12 days compared to the control samples. By the end of storage, tomatoes coated using the air-gun spraying method exhibited 7.27% weight loss, firmness at 5.44 MPa, 3.00 °Brix total soluble solids, 2.90% vitamin C, and a color $\Delta E \sim 1.84$, which overall outperformed those treated by dipping (weight loss: 7.42%; firmness: 6.32 MPa; TSS: 3.00 °Brix; vitamin C: 1.76%; and color ΔE: ~ 1.07) and manual air-pressure spraying (weight loss: 11.78%; firmness: 5.24 MPa; TSS: 3.90 °Brix; vitamin C: 1.58%; and color ΔE: ~ 1.62) (see Table S3 in the SI).

These differences are linked to coating thickness uniformity and gas-moisture regulation capability. Dipping produced thicker coating accumulation in certain areas, leading to anaerobic zones and tissue softening. Manual air-pressure spraying provided thinner but less uniform coatings, resulting in variability across samples. In contrast, air-gun spraying provided finer and more consistent atomization, a continuous and smoother layer, better automation, reduced material waste, and improved process control. Its coating uniformity also contributed to enhanced barrier properties. Therefore, air-gun spraying has been identified as a robust method to realize layer-by-layer applications on an industrial scale to extend the shelf-life of horticultural products. To contextualize these findings, a comparative summary with recent tomato-coating studies is provided in Table S4 in the SI. This comparison emphasizes the key advances of the current work, particularly in demonstrating that the application method plays a decisive role in coating uniformity, barrier functionality, and shelf-life enhancement. Overall, while most previous studies relied solely on static immersion, this study introduces a novel comparison of three application methods and demonstrates that spraying, particularly air-gun spraying, provides similar physical and quality improvement and achieves a longer shelf-life duration compared to some bioactive-enriched formulations. This highlights the practical applicability and process efficiency of the present work.

Despite the successful demonstration of the effective chitosan/HPMC coating in extending the shelf-life of tomatoes, the effects of particle size modification in the mixed solutions

on the coating characteristics are of interest for feasibly improving the application coating performance. Here, detailed particle analysis is therefore needed, where different material characterization methods (*e.g.*, dynamic light scattering (DLS)) can be conducted to determine the particle size distributions.[18,20,55,65,77,78] Additionally, for further study, microbial analysis is considered an important action for post-harvest tomato preservation to provide more quantitative data on possibly detected microorganisms, besides the physicochemical and quality attributes of coated tomatoes. Future optimization regarding mechanical conveyance and continuous spraying integration will further enhance cost-efficiency, enabling scalable adoption in fresh-produce supply chains.

4 Conclusions

The development and application of polysaccharide-based layer-by-layer edible coatings have been successfully conducted using a combination of chitosan and hydroxypropyl methylcellulose (HPMC), which can significantly improve the quality and shelf-life of fresh tomatoes up to 12 days longer than their uncoated counterpart. Among three different application methods (*i.e.*, dipping, air-gun spraying, and manual air-pressure spraying), air-gun spraying has been proven to be the most effective, consistent, and practical method for implementing chitosan/HPMC layer-by-layer on fresh tomatoes. It can limit final weight loss to 7.27%, maintain firmness at 5.44 MPa, and retain ~68% of the total soluble solids after 24 days of storage. This method also has strong potential to be applied to other climacteric fruits with similar deterioration behavior such as papaya, mango, and eggplant due to their high respiration rate, sensitivity to water loss, and perishability under ambient storage.

Author contributions

Aulal Muna: conceptualization, investigation, formal analysis, visualization, writing – original draft, and writing – review & editing. Rizky Aflaha: investigation, visualization, writing – original draft, and writing – review & editing. Syahla Salsabila and David Rusliman: investigation. Kuwat Triyana: supervision, resources. Aditya Rianjanu: validation and writing – review & editing. Hutomo Suryo Wasisto and Condro Wibowo: conceptualization, supervision, validation, investigation, funding acquisition, resources, and writing – review & editing.

Conflicts of interest

All authors declare that they have no conflicts of interest.

Data availability

All data to support the findings are available within the article, supplementary information (SI) and cited references. Supplementary information: comprises information on physicochemical properties of hydroxypropyl methylcellulose (HPMC) and chitosan edible films with various concentrations, physicochemical properties of the selected chitosan/HPMC edible coating treatments for fresh tomatoes during 24 days of storage, visualization of edible coating effects on fresh tomatoes during 24 days of storage, and comparative summary between this work and the previous studies. Additional information can be provided by corresponding authors upon reasonable request. See DOI: https://doi.org/10.1039/d5fb00555h.

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