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Fabrication and characterisation of fish gelatine-tapioca starch composite film incorporated with sucrose ester surfactant, and its application on shelf-life extension of cherry tomatoes

[1]*Fazial, F. F., [1]Teh, K. L., [2]Ahmad, A. A. and [1]Madzuki, I. N.

[1]Faculty of Chemical Engineering and Technology, Universiti Malaysia Perlis, Uniciti Alam Campus, 02100 Padang Besar, Perlis, Malaysia
[2]Green Biopolymer, Coating and Packaging Cluster, School of Industrial Technology, Universiti Sains Malaysia, 11800 Penang, Malaysia

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Abstract

In the present work, the incorporation of hydrophobic sucrose ester (SE) surfactant at various concentrations (5 to 20%, w/w) biopolymer into fish gelatine-tapioca starch composite film resulted in several significant changes. Specifically, there was a decrease in tensile strength (TS) from 14.79 to 7.18 MPa, an increase in water barrier properties from 0.9 to 0.6 g mm/m^2.h.Pa × 10^{-5}, an increase in the contact angle of the film from 66° to 78°, and enhanced opacity when the SE concentration was increased from 5 to 20% (w/w) biopolymer. The present work is novel in its approach to incorporating a hydrophobic SE surfactant into a composite biopolymer-based film, which has not been extensively explored for its combined effects on mechanical, water barrier, and shelf-life extension properties in food packaging applications. Wrapping cherry tomatoes with this SE-incorporated composite film slowed the degradation processes, such as decreased redness intensity, weight loss, firmness, and acidity titratability, likely due to the film's effectiveness in preventing moisture accumulation, which can accelerate bacterial and mould spoilage. The findings of the present work suggested that including SE as a packaging component could mitigate the hydrophilic nature of protein-carbohydrate-based films, thereby enhancing their effectiveness. The film's improved resistance to external water molecule penetration suggested its potential as an alternative packaging solution for various food products.

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Introduction

The advantages and unique characteristics of edible films over synthetic food packaging have driven a significant rise in alternative packaging materials in recent years. Edible films, especially when enhanced with beneficial components during production, offer promising potential for improving food preservation and sustainability. In contrast, synthetic plastics, which are produced at a staggering rate of 200 million tons per year (Jambeck *et al*., 2023), are made from thermoplastic polymers such as polystyrene, polyolefin, and polyvinyl chloride (PVC). These materials often take centuries to decompose, which negatively impacts the environment, and creates more waste disposal issues (Oghly, 2023).

Among the types of edible film, protein-carbohydrate composites have garnered attention due to their capability to form networks with enhanced functional properties. For instance, the combination of chicken skin gelatine and tapioca starch was the subject of the only work to date on gelatine-tapioca composite film for food packaging (Loo and Sarbon, 2020). However, protein-based films often exhibit high water vapour permeability due to the polarity of hydrophilic amino acids, thus limiting their application (Fakhoury *et al*., 2012). Strategies to mitigate this issue include the addition of zinc oxide nanoparticles to improve water vapour control (Lee *et al*., 2021). Another promising approach is the incorporation of lipid-based components like sucrose esters (SE). Sucrose esters which are derived from sugar and fatty acids, serve as emulsifiers in various

*Corresponding author.
Email: farahfaiqah@unimap.edu.my

industries including food, medicine, and cosmetics. Their hydrophilic-lipophilic balance (HLB) value which is determined by the fatty acid composition and degree of esterification, dictates their behaviour: high HLB values (8 - 18) indicate hydrophilicity, while low HLB values (4 - 6) indicate hydrophobicity (Zhang *et al*., 2023a). This makes SE with low HLB values particularly suitable as hydrophobic agents in edible films.

Sucrose esters have been incorporated into various natural polymeric matrices, including fish gelatine, arabinoxylan, pullulan, and hydroxypropyl methylcellulose (Amalini *et al*., 2018). Sucrose esters with HLB value of 5 have proven effective as coating agents for tomatoes by reducing water loss, lowering respiration rates, and maintaining quality during storage (Zhao *et al*., 2023). Despite their widespread use, the incorporation of SE as hydrophobic surfactants in fish gelatine-tapioca starch films for food packaging has yet to be fully explored.

The hydrophobic nature of SE can act as a barrier against water vapour transmission, and provide resistance to vapour and gas transport. Previous studies have shown the benefits of SE in extending the shelf life of fresh produce. Sucrose esters incorporated into rice starch film extended the post-harvest ripening of Cavendish bananas at 20°C by reducing the weight loss, prolonged the shelf life by up to 12 days, and delayed chlorophyll degradation (Thakur *et al*., 2019). Additionally, chicken packaging incorporated with fatty acids improved the barrier properties of films against water and light, slowing the degradation of vitamin C (Wu *et al*., 2023).

Cherry tomatoes, being perishable fruits, are prone to fungal infections, especially in humid conditions, which can accelerate decay and contribute to spoilage. Developing hydrophobic packaging to control moisture and decay is a crucial strategy for maintaining the quality of cherry tomatoes, reducing food waste, and enhancing consumer satisfaction (Korte *et al*., 2023).

The present work thus evaluated the incorporation of SE into a fish gelatine-tapioca starch composite film at varying concentrations. The growing demand for fish gelatine and tapioca starch is due to their attractive properties of abundance, low cost, biodegradability, and film-forming abilities (Siddiqui *et al*., 2023). By incorporating SE into this composite, we aimed to enhance the film's moisture and gas barrier properties, which could extend the shelf life of cherry tomatoes by preventing dehydration and oxidative processes. This innovative approach has the potential to significantly improve the preservation of fresh produce, marking a novel contribution to the field of sustainable food packaging.

Materials and methods

Materials and reagents

Fish gelatine from black tilapia skin and tapioca starch were obtained from a local supplier at Kangar Perlis, Malaysia. Sucrose ester with HLB-5 was obtained from Liuzhou Aigefu Food Technology Co., Ltd. (Liuzhou, China). Glycerol, magnesium nitrate ($MgNO_3$), citric acid, and glycerol were obtained from Merck Co. (Darmstadt, Germany).

Preparation of film-forming solution

The film-forming solutions were prepared using a series of mixtures of 5% tapioca starch and 5% fish gelatine at a ratio of 75:25 with the incorporation of SE at 5 to 20% (w/w, polymer). Each mixture was made up to the total weight basis (80 g), which included 10% (w/w, polymer) of glycerol in distilled water. The 5% tapioca starch was dissolved in distilled water, and heated in a water bath with the presence of stirrer using a magnetic hotplate at 85°C for 30 min until completely gelatinised. The fish gelatine was dissolved at 60°C for 30 min in distilled water until a clear solution was obtained. The fish gelatine solution was then added to gelatinised tapioca starch at 60°C, and stirring was continued for 30 min. The mixture was incorporated with SE while constant stirring continued for another 30 min. Prior to film casting, the mixture was cooled to room temperature (Al-Hassan and Norziah, 2012). All samples were then labelled as SE0, SE5, SE10, SE15, and SE20 corresponding to the film forming solution containing 0, 5, 10, 15, or 20% SE (w/w, polymer), respectively.

Preparation of film by casting technique

The film-forming solutions were casted onto polyacrylic plates (16 cm × 16 cm × 3 mm) followed by oven-drying at 55°C for 4 h. Drying then continued at room temperature (25°C) for 24 h (Abedinia *et al*., 2021). The dry films obtained were peeled off and stored in a desiccator containing

saturated magnesium nitrate solution, Mg $(NO_3)_2$, at a relative humidity of 55% at room temperature for further analysis.

Fourier-transform-infrared spectroscopy (FTIR)

The film samples were dried in a desiccator with silica gel for 2 w before analysis. The resulting spectra were in the range of 4000 - 650 cm^{-1}, with 32 scans and 4 cm^{-1} resolution as recorded by Attenuated Fourier Transform Infrared spectroscopy (ATR-FTIR, Perkin Elmer Spectrum GX, Perkin Elmer, USA). The spectra obtained were used to analyse the interaction of the composite film with and without the incorporation of SE (Samani *et al*., 2023).

Physical and mechanical characterisation of film

The film thickness was measured using a digital micrometre (Mitutoyo, Japan). Ten different positions of the samples were randomly measured, and the average thickness was calculated.

Tests for tensile strength (TS), elongation at break (EAB), and Young's Modulus (E) were conducted following the ASTM D882-10 method (ASTM, 2018) with some modifications to evaluate the mechanical properties of the prepared film. Film specimen strips (7 × 1 cm) were cut and conditioned at room temperature for 48 h before being tested in a desiccator that contained saturated magnesium nitrate solution. To perform TS and EAB as a tension test, a TA.XT-Plus texture analyser (Stable Micro System, Surrey, UK) was used. It was equipped with a 5 kg load cell, and the crosshead speed was set at 1 mm/s. The samples were mounted between grips with an initial gap of 50 mm and film width of 1 cm. The results of TS, EAB, and E were expressed by MPa, percentages (%), and MPa, respectively. The test was repeated three times.

Light barrier properties

Light absorption and transparency of the films were measured according to Nikmanesh *et al*. (2023) with modification. The light barrier properties of the film were determined by exposing it to light absorption from wavelengths of 200 to 800 nm, with the absorbance at each wavelength recorded. To measure the film transparency, the film samples were cut into 4 × 1 cm strips, placed into a quartz cuvette, and directly inserted into a spectrophotometer test cell. An empty cuvette was used as blank. A UV-vis spectrophotometer (model UV-160, Shimadzu, Kyoto, Japan) was used to record the absorbance. The transparency (T) value of the film at 600 nm was calculated using Eq. 1:

$$T = \frac{A_{600}}{x} \quad \text{(Eq. 1)}$$

where, A_{600} = absorbance at a wavelength of 600 nm, and x = film thickness (mm). Based on Eq. 1, a greater value of T would represent a lower transparency of film. The test was repeated three times.

Water contact angle

The surface hydrophobicity of the film was reflected using water contact angle. This was measured using a Contact Angle Analyzer (CAM120, Creating Nano Technologies Inc., Tainan, Taiwan). The films were cut into 4 × 4 cm squares, and 10 µL drop of distilled water was deposited onto the surface of each film sample at room temperature. An image of the water droplet on the film surface was immediately captured by the camera with viewpoints perpendicular to each other. The test was repeated three times.

Water vapour permeability (WVP)

The water vapour permeability was measured following the ASTM E96-95 method with some modifications (ASTM, 1995). Cup containers with a diameter of 3.8 cm and a height of 2.8 cm were used. Initially, glass permeation cells containing 10 g of silica gel (0% RH) were oven-dried at 120°C for 1 day. The test films were sealed on top of the permeation cups using Parafilm. These test cups were placed in a desiccator that contains distilled water (100% RH). The test cups were weighed at intervals of 24 h over a seven-day period. The weight of the cups was recorded to the nearest 0.0001 g, and a graph of function of time was plotted. Linear regression ($r^2 \geq 0.99$) was used to calculate the slope of each line. The WVP of the films was calculated using Eq. 2:

$$WVP = \frac{WVTR \times L}{\Delta P} \quad \text{(Eq. 2)}$$

where, WVTR = water vapour transmission rate ($g\ m^{-2}h^{-1}$) through a film which was calculated using the slope of the straight line divided by the exposed film area (m^2), L = mean film thickness (mm), and ΔP = partial water vapour pressure difference (Pa) across the two sides of the film. The test was repeated three times.

Analysis of colour, weight loss, firmness, and titratable acidity of cherry tomatoes wrapped with selected film

Based on the results obtained, the formulation of fish gelatine-tapioca starch with 10% SE was selected to be applied on cherry tomatoes due to its excellent mechanical properties, barrier characteristics, protection against UV light, and high degree of contact angle. The wrapping method was conducted according to Farhan and Norziah (2020). The colour changes, weight loss, and firmness decrement of cherry tomatoes were observed and recorded for 10 d at 4°C to monitor the quality rate changes during the storage period. The cherry tomatoes were washed thoroughly and left to air-dry before being wrapped with the prepared film. The parameters of colour, weight loss, texture, and TA were determined every 2 d throughout the storage period (day 0, 2, 4, 6, 8, and 10). The sample at day 0 was the control sample due to the absence of SE in the composite film.

The redness of the cherry tomatoes was measured using a CR-400 Chroma Meter (Konica Minolta, Osaka, Japan). Each cherry tomato sample was measured six times. Cherry tomato's redness is an important parameter in determining its level of maturity as a fruit that is rich in lycopene (Pangesti *et al*., 2015).

Weighing method was used to measure the weight loss of cherry tomatoes. The same cherry tomatoes were weighed from the first day until the end of the experiment, and a weight reading was taken every 2 d during storage. The percentage loss compared to the initial total weight was expressed as the resulting weight loss. The initial weight before storage (Wo) was compared with the final weight after storage throughout the period to observe the differences. The percentage of weight loss was calculated using Eq. 3:

$$\text{Weight Loss } (\%) = \frac{\text{Final weight} - \text{Initial weight}}{\text{Initial weight}} \times 100 \quad \text{(Eq. 3)}$$

A TA.XT-Plus texture analyser was used to determine the firmness of the cherry tomatoes using the compression force. The tests were conducted with a 50 mm diameter cylindrical probe and a 50 N load. The test speed was set at 2 mm/s for the pre-test, and 5 mm/s for the post-test. The results were expressed in Newtons (da Costa de Quadros *et al*., 2020).

The titratable acidity (TA) of the cherry tomatoes was determined using the titration method described by Al-Dairi *et al*. (2021). About 5 mL of juice from the cherry tomatoes was taken and diluted using 95 mL of distilled water, with phenolphthalein as the indicator. The TA of cherry tomato juice was calculated by titrating 5 mL of cherry tomato juice against 0.1 N NaOH. The TA was expressed as a percentage of citric acid using Eq. 4, where 0.064 was the citric acid milliequivalent factor:

$$Titratable\ acidity\ (\%\ TA) = \frac{vol{:}NaOH(mL) \times 0.1\ (normality\ of\ NaOH) \times 0.064}{ml\ juice\ or\ g\ juice} \times 100 \quad \text{(Eq. 4)}$$

Statistical analysis

IBM SPSS 26.0 (USA) was used to analyse the data obtained in the present work. The film thickness, light barrier properties, mechanical test, water vapour permeability, water contact angle, colour, and texture data were subjected to One-way variance analysis using Duncan's test, with a confidence level of $p \leq 0.05$.

Results and discussion

FTIR analysis of composite film with SE

Spectra profiles of the fish gelatine-tapioca starch composite film with SE incorporated is shown in Figure 1. Generally, all functional groups representing the structure of gelatine and starch were observed in the spectra. The broad band observed at 3300 cm^{-1} indicated the stretching vibrations of the hydroxyl (—OH) group, which are present in both tapioca starch and fish gelatine. This band represents the intermolecular hydrogen bonding between these polymers, indicating the presence of a cohesive network structure within the film matrix (Luo *et al*., 2022). The band around 1600 cm^{-1} (Amide I) indicated a carbonyl group (C=O) in the peptide bonds of the gelatine which is a crucial indicator of the protein's secondary structure (Muyonga *et al*., 2004). The band around 1510 cm^{-1} (Amide II) indicated the stretching vibration of N—H group, while band around 1310 cm^{-1} (Amide III) indicated the C—N stretching vibrations in gelatine (Luo *et al*., 2022). Together, these bands confirmed the presence of gelatine's structural components in the composite film. The band around 1000 cm^{-1} indicated the C—O vibrations in gelatine and starch, further

demonstrating the successful blending of these two biopolymers in the film (Silva *et al*., 2019). The incorporation of SE into the composite film could be justified based on the presence of a band around 798 cm^{-1} which indicated SE. Furthermore, the increment of SE in the composite film caused the Amide B of the SE incorporated in the film (2923.2 cm^{-1}) to shift to lower frequencies compared to the control composite film (2934.5 cm^{-1}) (Amalini *et al*., 2018). This observation was due to the conformational changes and hydrophobic interaction between gelatine and SE (Zhang *et al*., 2023b). The incorporation of SE into the film produced a slightly noticeable band around 2854 cm^{-1}, which indicated the long carbon chain of SE. This finding was supported by Amalini *et al*. (2018), who also observed a noticeable band at 2850 cm^{-1} after the incorporation of fish gelatine film. Another work conducted by Yan *et al*. (2023) also showed a distinctive peak at 2850 - 2960 cm^{-1} due to the addition of hydrocarbon segments from fatty acids into the film matrix. This further confirmed the presence of SE in the composite film in the present work.

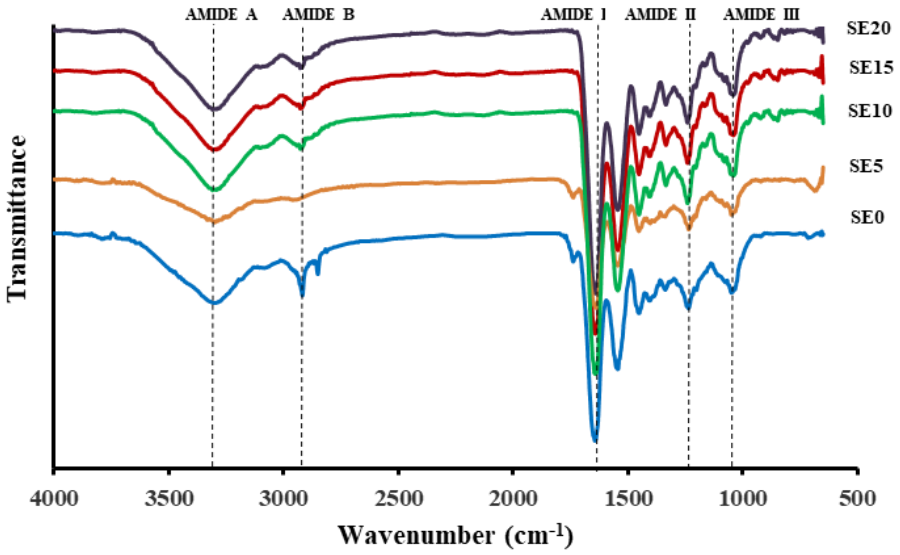

Figure 1. FTIR spectra of fish gelatine-tapioca starch composite film incorporated with different concentrations of sucrose ester (SE).

Physical and mechanical characterisations

Table 1 shows the physical and mechanical properties of the films prepared with different concentrations of SE. The film thickness significantly increased from 0.225 to 0.250 mm as SE concentration increased from 0 to 20%. This increase in thickness was primarily due to the interaction between SE and peptide chains which disrupted the ordered alignment and network arrangement (Tongnuanchan *et al*., 2013). This disruption was caused by the oxidation of fatty acids, resulting in the formation of radicals, lipid hydroperoxides, and other secondary lipid oxidation products (Ahotupa, 2024). These compounds covalently modified the side chains and polypeptide backbone of protein molecules, further contributing to the disruption within the film (Theerawitayaart *et al*., 2019).

The tensile strength (TS), elongation at break (EAB), and modulus of elasticity (E) of the composite films containing SE are also detailed in Table 1. With an increasing SE concentration (5 to 20%), both TS and E decreased significantly from 17.01 to 7.18 and 18.43 to 3.88 MPa, respectively. Conversely, EAB increased markedly from 94.35 to 185.44% with the incorporation of SE. This enhanced EAB was attributed to lipid plasticisation effects in the fish gelatine-tapioca starch composites, which facilitated the film extensibility (Said *et al*., 2023).

The primary interactions in the films involved hydrogen bonds between the polar hydrophilic heads of the SE, and the polar amino groups of gelatine, as well as hydrophobic interactions between the non-polar tails of SE, and the exposed hydrophobic amino groups of gelatine (Amalini *et al*., 2018). These interactions reduced protein-protein hydrogen bonding, altering the matrix orientation to include a hydrophobic SE phase, thus enhancing film flexibility. In contrast, control films without SE

exhibited less extensibility as the strong polysaccharide and protein hydrogen bonding hindered chain slippage during tensile testing (Tadele *et al*., 2023).

The decrease in TS with the increased concentration of SE can be explained by the dispersion of fatty acid droplets within the film network. This interfered with polymer interactions, and led to discontinuity in the film matrix. As the continuity of the film matrix was reduced, the cohesive structural integrity of the network in the film was compromised, thus lowering the TS (Tongnuanchan *et al*., 2015). Consequently, decreased TS correlated with decreased SE, indicating a less rigid film. The results of the present work demonstrated better mechanical properties than a previous work on basil seed gum edible films using different plasticisers and SE (Amini and Razavi, 2020). The TS and EAB values obtained in the present work, ranging from 5.7 to 9.9 MPa and 25.8 to 37.1%, respectively, were higher than those obtained in the previous work. This improvement might have been due to the use of polymer blends rather than a single polymer type, thus enhancing film properties.

Table 1. Film thickness and mechanical properties of fish gelatine-tapioca starch composite film incorporated with different concentrations of sucrose ester (SE).

Sample	(% SE, w/w, biopolymer)	Film thickness (mm)	Tensile strength (MPa)	EAB (%)	Young's Modulus, E (MPa)
SE0	0	0.225 ± 0.011^{d}	17.01 ± 0.48^{a}	94.35 ± 19.38^{b}	18.43 ± 2.96^{a}
SE5	5	0.231 ± 0.004^{cd}	14.79 ± 1.04^{b}	83.33 ± 4.81^{b}	17.78 ± 1.21^{a}
SE10	10	0.237 ± 0.008^{bc}	11.53 ± 2.11^{c}	99.34 ± 9.29^{b}	11.55 ± 1.04^{b}
SE15	15	0.240 ± 0.002^{b}	8.07 ± 0.38^{d}	163.78 ± 5.91^{a}	4.94 ± 0.26^{c}
SE20	20	0.250 ± 0.005^{a}	7.18 ± 0.90^{d}	185.44 ± 26.62^{a}	3.88 ± 0.07^{c}

Different lowercase superscripts within similar column indicate significant difference ($p \leq 0.05$).

Light transmission and transparency of film

In addition to the physical and mechanical properties of the film, favourable light transmission characteristics are crucial when designing food packaging. Typically, films made from gelatine-starch compositions exhibit moderate transparency due to the light scattering during the retrogradation of starch (Li *et al*., 2023). As shown in Table 2, as the SE concentration increased, the light transmission of the composite film decreased, resulting in reduced transparency. This indicated that the film became more opaque when the SE content was higher. However, these changes were not statistically significant. Similar results were obtained in a work on *Salvia macrosiphon* gum-based edible film, which indicated that an increase in SE content increased film opacity (Amini and Razavi, 2020).

This result was likely due to the alteration of the refractive index throughout the films, caused by the interaction between gelatine and fatty acid in the SE (Amalini *et al*., 2018). Additionally, increasing the SE concentration limited the water absorption into the film. Both the water and crystalline content affected the transparency of the film. Thus, the incorporation of SE decreased the water absorption into the films, resulting in changes in the molecular organisation of tapioca starch and fish gelatine, thereby increasing the transparency value of the film (Fakhouri *et al*., 2018).

Table 2. Light transmittance of fish gelatine-tapioca starch film incorporated with different concentrations of sucrose ester (SE).

Sample	Light transmission (%)					Transparency value (T_{600})
	400	500	600	400	500	
SE0	61.52 ± 0.81^{a}	72.00 ± 0.91^{a}	76.27 ± 0.44^{a}	78.28 ± 0.63^{a}	80.17 ± 0.18^{a}	0.56 ± 0.04^{c}
SE5	41.83 ± 2.38^{b}	59.18 ± 1.99^{b}	67.62 ± 1.76^{b}	72.07 ± 1.53^{b}	75.17 ± 1.19^{a}	0.64 ± 0.02^{c}
SE10	44.18 ± 1.88^{b}	61.69 ± 2.38^{b}	67.74 ± 2.47^{b}	71.07 ± 0.99^{b}	75.10 ± 0.10^{a}	0.74 ± 0.10^{c}
SE15	19.40 ± 0.52^{c}	27.65 ± 0.22^{c}	33.60 ± 0.66^{c}	37.54 ± 1.26^{c}	41.67 ± 1.41^{b}	2.20 ± 0.19^{b}
SE20	14.03 ± 3.73^{d}	21.16 ± 0.71^{d}	21.99 ± 5.54^{d}	25.68 ± 6.59^{d}	26.76 ± 6.45^{c}	2.87 ± 0.50^{a}

Different lowercase superscripts within similar column indicate significant difference ($p \leq 0.05$).

Water contact angle

Contact angle measurements provide valuable insights into the suitability of packaging materials for different food products. A high-water contact angle is desirable for enhancing the moisture resistance of films. Figure 2a displays the water contact angle measurements for the composite films with varying concentrations of SE (5 to 20%). An increase in the amount of SE, a hydrophobic material, led to a gradual increase in the contact angle of the composite films, attributable to the hydrophobicity of SE. The contact angle significantly increased from 66.02° to 76.52° when the SE concentration was increased from 0 to 15%. However, beyond a 15% increase to 20%, no significant difference was observed. This could be because at higher SE contents, a small portion of the lipid phase tended to migrate from the lower surface to the air interface, influenced by the density difference between the two phases (Wang *et al*., 2024). This result was in agreement with a previous finding that with a higher olive oil content at 15%, there were no significant differences in the contact angle (Pereda *et al*., 2012).

The increase in the contact angle with the incorporation of SE was due to the interaction between the hydrophilic groups of SE molecules and the polymer chains of fish gelatine-tapioca starch. This interaction repelled the aliphatic side chains of

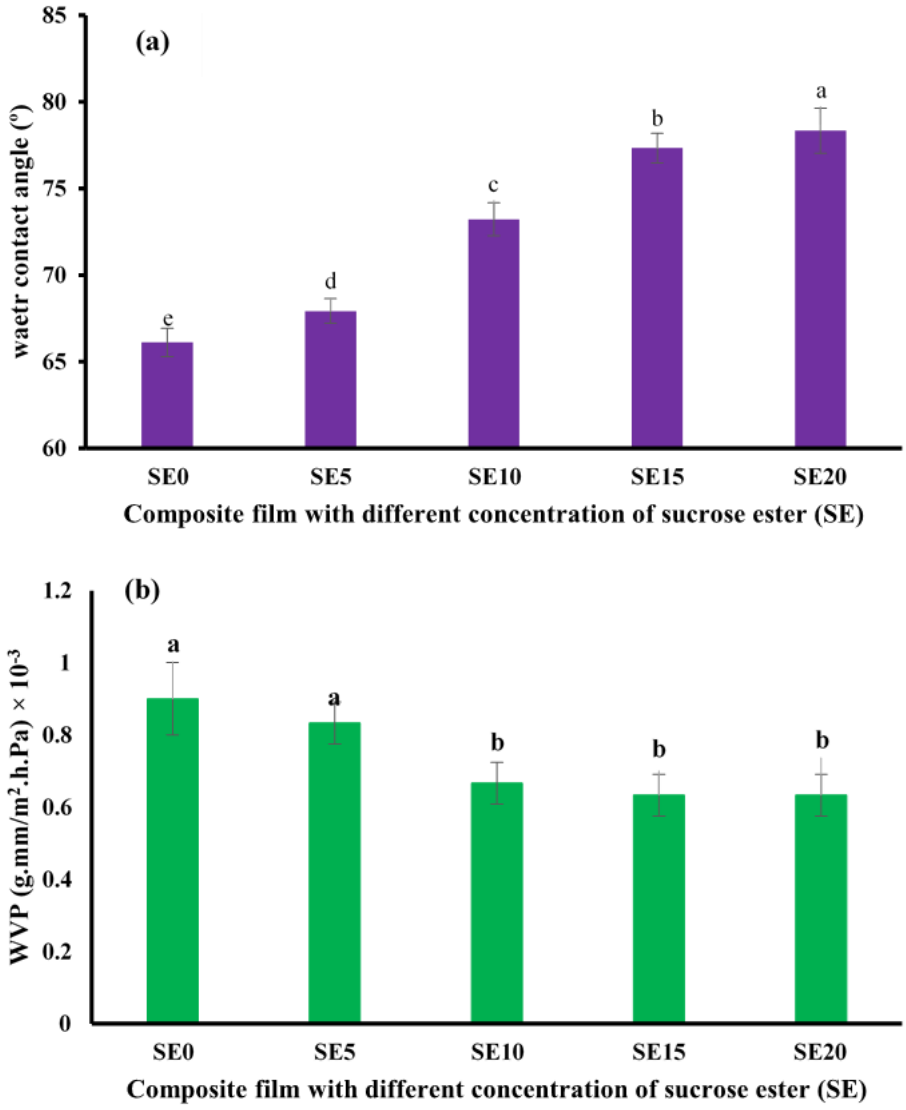

Figure 2. Water contact angle **(a)** and water vapour permeability (WVP) **(b)** of fish gelatine-tapioca starch composite film incorporated with different concentrations of sucrose ester (SE). Different lowercase letters indicate significant difference ($p \leq 0.05$).

the SE to the exterior parts of the film, making it more hydrophobic. Additionally, the increase in the contact angle with increasing SE concentration was due to the dispersion of hydrophobic SE in the solution, which reduced the availability of hydroxyl groups to interact with water. Consequently, this led to a less moisture absorption by the film (Ma *et al*., 2016).

The incorporation of SE may immobilise the biopolymer chains, thus reducing the interaction tendency with water molecules. The contact angle value was better than what Amini and Razavi (2020) obtained, with their work producing values of 70.27°, 68.69°, and 69.51° when starch-gelatine film was incorporated with palmitic, stearic, and oleic acids, respectively. This discrepancy could be due to the different biopolymers used to form the composite film; the previous work by Amini and Razavi (2020) used *Salvia macrosiphon* gum, whereas the present work employed fish gelatine and tapioca starch.

Water vapour permeability (WVP)

One of the main functions of food packaging is to minimise moisture transfer between the food and the surrounding atmosphere. The penetration of water into packaging film can reduce the water contact angle, as mentioned in the previous section, leading to a wetter film, and an increased risk of food spoilage. Therefore, it is essential to keep the water vapour permeability (WVP) of the film as low as possible (Adjouman *et al*., 2017). Based on Figure 2b, a significant decrease in WVP value was observed as the SE concentration increased from 5% (8.64×10^{-4} ± 0.58 g mm/Pa h m^2) to 10% (7.13×10^{-4} ± 0.96 g mm/Pa h m^2). However, further increases in SE concentration did not result in a significant decrease in WVP. This can be attributed to the disruption of the polymer matrix caused by the hydrophobic molecules in SE. At higher SE concentrations (15 and 20%), the decrease in the WVP value of the film became insignificant as these hydrophobic compounds had less compatibility with the matrix at high concentrations. At lower SE concentrations (around 10%), the disruption of the matrix did not occur, and the hydrophobic effect on the film was more pronounced (10% SE) (Fakhouri *et al*., 2018).

As the SE concentration increased, the water vapour transmission rate of the composite film gradually decreased, thus enhancing its moisture resistance. This was due to the hydrophobic properties of SE, which decreased the water vapour transmission rate, and limited the formation of hydrogen bonds within the composite film matrices (Wang *et al*., 2019). The presence of a hydrophobic dispersed phase in the film network created a tortuous path for water molecules to pass through, effectively decreasing the WVP of the film (Hashemi Gahruie *et al*., 2020). Additionally, the hydrophobic moieties of SE decreased the affinity of fish gelatine chains toward water vapour. The WVP value of the films was also affected by the degree of saturation, polarity, chain length, and branching of the SE molecules. Therefore, longer, non-polar, saturated, and linear lipid molecules produced films that were more rigid and less permeable compared to polar, short, unsaturated, and a highly branched lipid molecule (Hashemi Gahruie *et al*., 2020).

The WVP values obtained in the present work can be compared with those from a previous work on gelatine and gluten proteins edible film containing SE, where the WVP values ranged from 0.93 to 2.19 $\times 10^{-3}$ g mm/Pa h m^2 at SE concentrations of 5 and 15%. The lower WVP values obtained in the present work were likely due to the differences in the types of fatty acids used (palmitic, myristic, lauric, capric, caprylic, and caproic acids) (Fakhouri *et al*., 2018).

Performance of cherry tomatoes wrapped in SE composite film

Based on the earlier discussion, SE10 was found to possess excellent mechanical and barrier properties, making it suitable for application with cherry tomatoes. Additionally, incorporating SE in concentrations higher than 10% was deemed unnecessary, as this did not further decrease the WVP. Figure 3a shows the colour changes of cherry tomatoes wrapped in fish gelatine-tapioca starch film containing 10% SE when stored for 10 d at 4°C. Cherry tomatoes typically exhibit a vibrant red colour, indicative of their maturity and high antioxidant lycopene content. The *a** value, representing the redness of the control cherry tomatoes, increased significantly from 43.2 to 83%, while the SE-wrapped cherry tomatoes showed an increase from 36.87 to 71.67%. The increase in the red value was driven by the natural ripening process, characterised by the breakdown of chlorophyll, and the accumulation of lycopene, which are promoted by ethylene production during storage (Kapoor *et al*., 2022). However, the red colour was more intense in the control cherry tomatoes compared to the SE-

wrapped cherry tomatoes. This result suggested that the SE-film could form a protective barrier that slows or prevents the degradation of lycopene when exposed to heat and light (Devilla *et al.*, 2015).

The percentage of weight loss of the control sample was higher than that of the SE-wrapped cherry tomatoes. The weight loss of the control and SE-wrapped cherry tomatoes increased to 11.6929 and 8.4331%, respectively. Higher weight loss could be seen in the control sample (Figure 3b). The weight loss was attributed to the respiration process, which involves the loss of carbon atoms in the form of carbon dioxide molecules leaving the cherry tomato surface, as well as the dehydration process through the transpiration of water molecules from the cells to the ambient atmosphere, which explained the increase in weight loss during storage (Sucharitha *et al.*, 2018). The hydrophobic nature of SE minimised the release of water molecules passing through the packaging film. Another perspective suggested that metabolic degradation in the cell wall caused the water retention capacity to decrease, as observed during senescence (Cipolatti *et al.*, 2012).

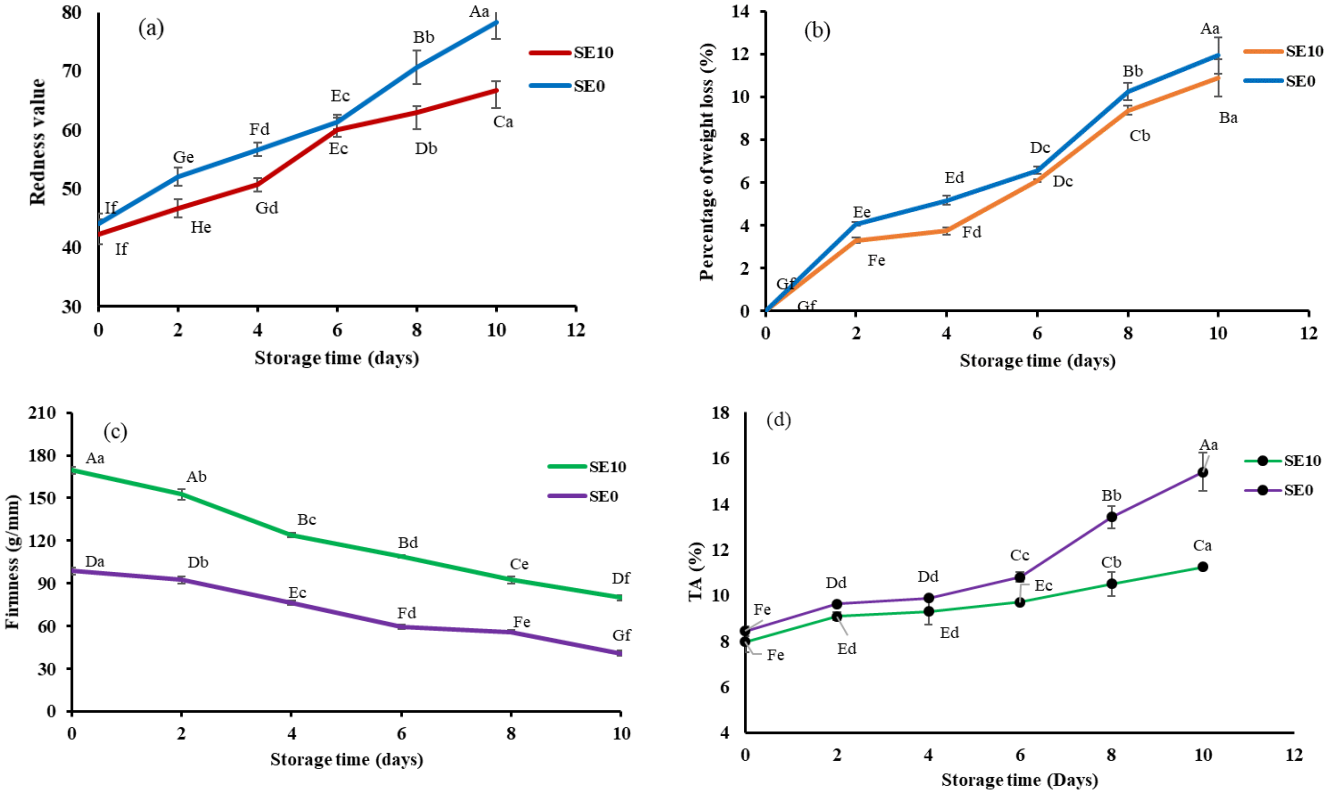

Figure 3. Comparison output of redness **(a)**, weight **(b)**, firmness **(c)**, and titratable acidity **(d)** changes of cherry tomatoes wrapped with fish gelatine-tapioca starch composite film, with and without incorporation of SE10, for 10 days observation at 4°C. Different uppercase letters indicate significant difference ($p < 0.05$) between samples. Different lowercase letters indicate significant difference ($p < 0.05$) between storage times.

Firmness reductions are often attributed to different factors such as a loss of cell turgor pressure as well as cell wall and polysaccharide degradation. Applying the SE composite film on the cherry tomatoes significantly ($p < 0.05$) reduced the firmness of the cherry tomatoes when stored at 4°C during experiment (Figure 3c). Initially, the cherry tomatoes had different degrees of firmness, with the cherry tomatoes wrapped in the control and fish gelatine-tapioca starch film with 10% SE having firmness levels of 187.71 and 173.85 g/mm, respectively. After 10 d of storage, the firmness of both the control and the 10% SE film wrapped cherry tomatoes had decreased to 131.93 and 140.71 g/mm, respectively. Compared to the control, the firmness loss for 10% SE film wrapped cherry tomatoes was less intense with a difference of 133.14 g/mm since the first day of storage. The results demonstrated that the applied film effectively extended the post-harvest life of cherry tomatoes by preserving their quality. This

protective layer can enable a more stable internal environment to be maintained, and slow the enzymatic reactions such as those of pectinase and cellulase, thereby preserving the structural integrity of the fruit, and slowing the softening process (Huang *et al*., 2023). The application of 2% of pineapple core carboxymethyl cellulose (CMCpc) and 2% of commercial carboxymethyl cellulose (CMCc) edible coatings on cherry tomatoes compared with control resulted in decrements in the firmness of cherry tomatoes of 481.31, 440.52, and 438.48 g/mm for control, CMCpc, and CMCc samples, respectively, after 20 d of storage (Pangesti *et al*., 2015). The firmness loss of the cherry tomatoes with a coating was less critical compared to that of the control.

The titratable acidity (TA) of the SE-wrapped cherry tomatoes during storage at 4°C increased from 8 to 11% during 10 d of storage (Figure 3d). The increase in the sour taste of cherry tomatoes was mainly because of the presence of citric and malic acids, and the "sourness" is closely related to the TA (Majidi *et al*., 2011). After day 4, the TA increment was lower than that of the control sample. This indicated that the composition and permeability of the fish gelatine-tapioca starch composite film may impact the oxygen and carbon dioxide gas exchange, which can influence the respiratory activity of the cherry tomatoes and the production of organic acids, thus impacting the TA during storage (Matloob *et al*., 2023).

Conclusion

The modification of films with sucrose ester (SE) successfully enhanced the surface hydrophobicity, barrier properties, and mechanical performance of fish gelatine-tapioca starch composite films. The water contact angle increased by 2° to 12°, the water vapour permeability (WVP) improved, and elongation at break (EAB) significantly increased from 94 to 185% when using SE concentrations ranging from 5 to 20%. Among the formulations, SE10 demonstrated optimal mechanical strength and moisture barrier properties, making it a promising candidate for food packaging applications. Beyond 10% SE, no significant improvement in WVP was observed, indicating a concentration limit for achieving the maximum benefits. When applied to cherry tomatoes, SE10 confirmed its capacity to maintain film stability and improve protective properties. However, limitations emerged, particularly with regard to the lack of significant improvements in WVP at higher SE concentrations. Further research should explore the microstructural interactions between SE and biopolymers using imaging techniques like microscopy, which could reveal the underlying mechanisms behind these changes. Additionally, testing the film's effectiveness against mould growth in a broader range of fruits could provide a more comprehensive understanding of its potential as an alternative to synthetic packaging. Despite these challenges, the modified films present a sustainable option for reducing reliance on synthetic polymers in food packaging, thus contributing to environmentally friendly solutions.

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