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Tapioca starch-based films containing oregano, Vietnamese mint, and curry leaf essential oils for food packaging applications

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Abstract

Sustainable food packaging made from starch is a viable alternative to the usage of petrochemical-based plastics. The incorporation of additives, particularly essential oils, into the starch film can potentially improve the mechanical properties and enhance its functionality. The present work investigated the effects of the incorporation of different types and concentrations (0, 0.25, 0.5, 0.75, and 1%; v/v) of essential oils (EOs) from oregano, Vietnamese mint (VEO), and curry leaf on the mechanical and optical properties of starch films. Additionally, the quality attributes of strawberries wrapped using the films during storage were investigated in terms of weight loss, colour changes, and firmness. It was observed that the incorporation of EOs into the starch films improved the elongation at break, especially at higher concentrations. These films became slightly yellowish and more opaque. Besides, the changes in quality attributes of strawberries wrapped with starch/EOs films were the lowest as compared to the unwrapped strawberries and the strawberries wrapped with starch films. Starch films incorporated with 1% (v/v) VEO were the most effective films due to relatively high flexibility and low stiffness, apart from the ability to decrease the changes in quality attributes of strawberries. To conclude, the starch/EOs films have great potential for food packaging applications.

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Introduction

The use of biopolymers in food packaging applications to replace some synthetic plastic usages and reduce environmental pollution is very promising due to their availability, low-cost production, biodegradability, and origin from renewable sources (Othman *et al.*, 2021). Biopolymers can also be used as carriers for additives such as antioxidant and antimicrobial agents, vitamins, flavours, and pigments that act as compound-releasing packaging to help improve the quality, and increase the shelf-life of the foods (Nordin *et al.*, 2020). Among the many types of biopolymers, starch is one of the most abundant natural examples. Considering its low cost, degradability, edibility, non-toxicity, and ease of chemical modification, many studies have focused on designing starch for film applications (Santos *et al.*, 2014; Šuput *et al.*, 2016; Nordin *et al.*, 2020; Othman *et al.*, 2021). In the present work, tapioca starch was investigated for film applications due to its suitable film-forming property, and exhibits appropriate physical characteristics, since the films are odourless, tasteless, colourless, and impermeable to oxygen (Chillo *et al.*, 2008). Moreover, tapioca plant also has a high tolerance for poor climate conditions, thus providing a good supply of starch to produce food packaging materials (Hanif *et al.*, 2016).

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However, starch-based films exhibit several drawbacks such as poor mechanical and barrier properties that limit their application in the food packaging industry. The film made from starch is generally brittle. For wrapping food, it is essential to produce a rigid and flexible structure of films (Othman *et al.*, 2019). Incorporating additives such as plasticiser, filler, as well as antibacterial and antioxidant agents can enhance the properties of the films. For example, the flexibility of the films can be improved with the incorporation of plasticisers, which increases the elongation at break of the films. However, the incorporation of plasticiser produces hygroscopic films with high water vapour permeability, thus causing the films to exhibit low barrier properties as it absorbs moisture from the atmosphere (Santos *et al.*, 2014). This limitation can be improved by incorporating hydrophobic components, particularly lipids into the films (Caetano *et al.*, 2017).

Essential oils (EOs) are one of the interesting lipids that exhibit antimicrobial and antioxidant capacities which can add functionality to the films. EOs are aromatic oily liquids obtained from plant materials, and useful in food packaging applications (Carpena *et al.*, 2021). EOs are a mixture of over 300 compounds, primarily consisting of volatile compounds with a low molecular weight of about 1,000 Da (Bhavaniramya *et al.*, 2019). Examples of EOs are from oregano, curry leaf, and Vietnamese mint. These EOs are recognised for their antimicrobial and antioxidant properties, which are useful and favourable in food industries (Carpena *et al.*, 2021).

EO of oregano exhibits antimicrobial, antioxidant, anti-inflammatory, antidiabetic, and cancer suppressor properties which are very beneficial in the food, cosmetic, and pharmaceutical industries (Leyva-López *et al.*, 2017). Curry leaf tree is a small strong-smelling perennial shrub commonly found in forests as undergrowth. The EO from the leaves is found to possess antioxidant, antibacterial, antifungal, larvicidal, anticarcinogenic, hypoglycaemic, anti-lipid peroxidative, hypolipidemic, and antihypertensive activities (Rajendran *et al.*, 2014). Vietnamese mint is a creeping, perennial herb. The EO from Vietnamese mint leaves contains aldehydes such as decanal and dodecanal, and the alcohol decanol (WikiDoc, 2011). Therefore, it is non-toxic and beneficial for food application to detoxify food. In addition, the EO from the leaves has also been reported to have antibacterial activity against Gram-positive and Gram-negative bacteria (Sasongko and Kerdchoechuen, 2011). The incorporation of EOs into the food packaging films will result in the production of functional packaging films that can help to extend the shelf life of food products (Carpena *et al.*, 2021).

Nevertheless, the types and concentrations of the EOs incorporated into the films are important parameters that can affect the properties of the films including the optical and mechanical properties (Pirouzifard *et al.*, 2019), which will affect the shelf life and quality attributes of the food products packaged with the films. Therefore, it is critical to determine the ideal type and concentration of the EOs. To the best of our knowledge, no work has been done to investigate the effects of the incorporation of different types and concentrations of EOs, particularly oregano, curry leaf, and Vietnamese mint EOs on the optical and mechanical properties of tapioca starch films or the quality attributes of the food products packaged with the films. Hence, the present work investigated the effects of different types and concentrations of EOs on the optical and mechanical properties of starch-based films. The potential application of the films as food packaging materials was demonstrated by analysing the quality attributes of strawberries wrapped with the films, and stored at 4°C, which included weight loss, colour changes, and firmness. The application of these EOs could provide new opportunities to develop a novel food packaging system that has acceptable performance and is safer for the environment.

Materials and methods

Tapioca starch (Brand Kapal ABC, Malaysia) was purchased from a local supermarket. Oregano (*Origanum vulgare*), Vietnamese mint (*Persicaria odorata*), and curry leaf (*Murraya koenigii*) EOs (Brand Soul) were purchased from BF1 Malaysia (Malaysia). Glycerol was purchased from R&M Marketing (UK). Strawberries (Cameron strawberry) were purchased from AEON Co. (M) Bhd. (Malaysia). All chemicals were used as received without further purification.

Film preparation

Tapioca starch (3 g) were dissolved in 100 mL of distilled-water-glycerol solution to obtain 3% (w/w) suspension starch solution. The concentration of glycerol used was 25% (w/w) of the dry starch solid weight. The starch solution was heated with continuous stirring using a magnetic stirrer hotplate (FAVORIT, PLT Scientific Sdn. Bhd., Malaysia) until the solution gelatinised at 75°C. After that, the solution was cooled down to 30°C. Then, different types (oregano (OEO), curry leaf (CEO), and Vietnamese mint (VEO)) and concentrations (0, 0.25, 0.5, 0.75, 1% (v/v) of film-forming solution) of EOs were incorporated into the respective solution with continuous stirring to produce starch/OEO0.25, starch/OEO0.5, starch/OEO0.75, starch/OEO1, starch/CEO0.25, starch/CEO0.5, starch/CEO0.75, starch/CEO1, starch/VEO0.25, starch/VEO0.5, starch/VEO0.75, and starch/VEO1 films. The films without EO were also prepared as control films. The concentrations for the EO were chosen based on the literature review of several works related to starch/EOs where the concentrations of EO used were in the range of 0.25 - 0.5 mL (Pirouzifard *et al*., 2019), 0 - 2% relative to the solution volume (dos Santos Caetano *et al*., 2018), 0.2 - 1% (w/v) of film solution (Cao *et al*., 2017), and 1 - 3% (v/v) of film solution (Ghasemlou *et al*., 2013). The samples were then subjected to ultrasonication (50% amplitude) for 5 min using an ultrasonic probe (QSonica, USA) to produce a homogenised solution. Then, 50 mL of film-forming solution was poured onto a Petri dish, and left to dry in an air-conditioned room (20°C) for 2 d. After drying, the film was peeled off from the Petri dish, and conditioned at 27°C and 54% relative humidity in an incubator (Memmert, Memmert GMBH +Co., Germany) before further analyses.

Film thickness

The thickness of the films was determined using a digital micrometre (Mitutoyo, Japan) at random positions on the films. The average value of the film's thickness was used to calculate the opacity, tensile strength (TS), elongation at break (EAB), and Young's modulus (YM) of the films.

Film optical properties

The optical properties of the films were determined using a colour spectrophotometer (Hunter lab, Ultrascan Pro, USA). L* (L = 100 (white), L = 0 (black)) represents lightness, a* represents green (+) to red (-), and b* represents blue (-) to yellow (+). These values were determined following the CIELAB colour parameters.

Eq. 1 was used to calculate the ΔE index which estimates the colour difference between samples:

$$\Delta E = \mathrm{sqrt}((L^*-L)^2 + (a^*-a)^2 + (b^*-b)^2) \quad \text{(Eq. 1)}$$

where, L*, a*, and b* = colour parameters of the starch/EO films, and L, a, and b = colour parameters of the starch films (control). The percentage of light transmittance of the films was recorded at the wavelength range of 200 to 800 nm. The opacity values of the films were calculated using Eq. 2:

$$\text{Opacity} = A600 / L \quad \text{(Eq. 2)}$$

where, A600 = transmittance value at 600 nm, and L = thickness of the film.

Film mechanical properties

The mechanical properties of the films were determined by measuring the TS, EAB, and YM of the films. These are the key indicators of film strength, flexibility, and stiffness, respectively. The film specimens were cut into strips of 100 × 15 mm after preconditioning. TS, EAB, and YM were determined using a texture analyser (TA.XT2 Stable Micro Systems, UK) equipped with a computer, following ASTM D882-02 method (ASTM, 2000). Film samples were mounted to the grip with an initial grip separation of 60 mm, and tensioned at 0.5 mm/s till break.

Quality attributes of strawberries

The strawberries were sorted to a uniform size, colour, and physical appearance (Shapi'i *et al*., 2020). The strawberries were selected at the stage where the fruits were in uniform red colour, and without any sign of mechanical damage or fungal deterioration. Then, the strawberries were washed with distilled water and wiped with a clean cloth. The strawberries were then individually wrapped with starch and starch/EOs films (Shapi'i *et al*., 2020) to ensure complete contact of the fruit with the film, and stored in a chiller (Protech, Tech Lab., Malaysia) at 4 ± 2°C storage temperature for 9 d (Del-Valle *et al.*, 2005). The changes in quality attributes of the strawberries in terms of weight loss, colour changes, and firmness were evaluated on days 0, 3, 6, and 9. For this purpose, each sample was unwrapped, and the following evaluations were conducted.

The determination of weight loss of the strawberry during storage was conducted using a weighing balance (Shimadzu, Shimadzu Corp, Japan). The weight loss percentage was calculated using Eq. 3:

$$\text{Weight loss (\%)} = (M_0 - M_i / M_0) \times 100 \quad \text{(Eq. 3)}$$

where, M_0 = weight of the strawberry on day 0, and M_i = weight of the strawberry on the sampling day.

Meanwhile, a precise colour reader (WR-18, Shenzhen Wave Optoelectronics, China) was used to examine the colour changes of the strawberries during the storage period. Calibration was performed using a white plate. To avoid the effects of heterogeneity on the fruit surface, the colour measurements were taken on the shoulder of each fruit. CIELAB colour parameters particularly L* (lightness) and a* (-greenness to + redness) were recorded. The a* value is the main index for fruit senescence.

The firmness of the strawberries was determined using a texture analyser (TA.XT2 Stable Micro Systems, UK) by measuring the force required for a 2 mm probe to penetrate 6 mm fruits at 1 mm/s. The samples were placed in such a way that the probe penetrated the fruits at the equatorial zone.

Statistical analysis

The statistical analysis of the measured experimental results was performed by analysis of variance (ANOVA) using Minitab 19 (Minitab 19, Minitab Inc., USA) software. The mean comparisons were conducted using Tukey's test at a 0.05 level of significance.

Results and discussion

In general, it was found that tapioca starch films containing different types and concentrations of EOs that were prepared *via* the solution casting method were peelable. The control film was found to be brittle, rigid, and the most transparent among others. The films containing EOs were more flexible, slightly yellowish, and exuded a light odour. The strawberries wrapped with films incorporated with EOs were found to have better quality attributes as compared to those wrapped with control films.

Film thickness

Table 1 shows that the incorporation of EOs into the starch films increased the thickness of the films slightly, with the increment being prominent at higher concentrations. The thickness of the films increased from 0.071 to 0.103, 0.115, and 0.099 mm ($p < 0.05$) as the control films were incorporated with 1% (v/v) OEO, CEO, and VEO, respectively. Nugroho *et al.* (2013) stated that the increase in film thickness was caused by the differences in the material concentration in the films due to the incorporation of different concentrations of EOs, even though the same volume of film-forming solution was poured onto the Petri dish during preparation. Therefore, the total solid contents of the films increased after the drying process due to the increase in film matrix constituent polymers.

Meanwhile, the thickness of starch/CEO1 films (0.115 mm) was greater than starch/OEO1 (0.103 mm) and starch/VEO1 (0.099 mm) films. This could have been due to the difference in material concentrations, as the CEO had higher total solid contents as compared to others as evidenced by the °Brix value. The °Brix value of CEO by hydrodistillation was 77.8 °Brix, which exhibited a dense fluid property (Tran *et al.*, 2012). Therefore, the incorporation of highly viscous CEO led to the increase in the thickness of the films. Meanwhile, the °Brix value of OEO was 76.1 °Brix (Pisoschi and Negulescu, 2012) and 72.7 °Brix for VEO (Sasongko and Kerdchoechuen, 2011), which were lower than CEO. The trend of film thickness observed in the present work followed the trend of the °Brix values.

Film optical properties

Colour, as well as the opacity of the packaging films, are important parameters that determine the consumers' acceptance of the films, and food products packaged with the films. The effects of incorporating EOs into the starch films on the L*, a*, b*, ΔE, and opacity are shown in Table 1. L* represents the lightness of the films from black to white. There was a slight decrease ($p < 0.05$) in L* values for all the starch films as the EOs were incorporated into the films, thus indicating a slight darkening of the films with EOs incorporation. The decrease became prominent at higher concentrations, especially at 1% (v/v). L* decreased from 92.91 to 89.36, 90.16, and 89.95 ($p < 0.05$) as the control films were incorporated with 1% (v/v) of OEO, CEO, and VEO, respectively. Nonetheless, the decrease was very small and almost unnoticeable when the films

Table 1. Thickness, L*, a*, and b* values of the films.

Starch film	Thickness (mm)	L*	a*	b*	ΔE	Opacity
Control	0.071 ± 0.002^{a}	92.91 ± 0.07^{a}	-0.74 ± 0.05^{a}	3.38 ± 0.04^{a}	0	0.79 ± 0.10^{a}
OEO0.25	0.077 ± 0.004^{Aa}	91.04 ± 0.42^{Ab}	-0.76 ± 0.04^{Aa}	3.51 ± 0.06^{Aab}	1.86 ± 0.86^{Aa}	1.21 ± 0.09^{Ab}
OEO0.5	0.078 ± 0.003^{Aa}	90.76 ± 0.42^{Abc}	-0.78 ± 0.06^{Aa}	3.53 ± 0.07^{Aab}	2.41 ± 0.86^{Aab}	1.27 ± 0.06^{Ab}
OEO0.75	0.093 ± 0.006^{Ab}	89.54 ± 0.83^{Acd}	-0.80 ± 0.03^{Aa}	3.77 ± 0.12^{Abc}	5.93 ± 2.77^{Abc}	1.34 ± 0.03^{Ab}
OEO1	0.103 ± 0.009^{Ab}	89.36 ± 0.46^{Ad}	-0.83 ± 0.02^{Aa}	3.85 ± 0.21^{Ac}	6.72 ± 0.55^{Ac}	1.45 ± 0.16^{Ab}
CEO0.25	0.081 ± 0.001^{Aa}	91.52 ± 0.57^{Aab}	-0.92 ± 0.09^{Aab}	3.67 ± 0.07^{Aab}	1.30 ± 1.03^{Aa}	0.85 ± 0.10^{Ba}
CEO0.5	0.099 ± 0.009^{Bb}	91.48 ± 0.89^{Ab}	-0.12 ± 0.21^{Bbc}	4.79 ± 0.76^{Bb}	2.01 ± 0.57^{Aa}	0.99 ± 0.07^{ABa}
CEO0.75	0.110 ± 0.007^{Ab}	90.49 ± 0.34^{Ab}	-1.25 ± 0.27^{Bbc}	4.85 ± 0.36^{Bb}	4.76 ± 0.79^{Aab}	1.16 ± 0.06^{Aab}
CEO1	0.115 ± 0.002^{Ab}	90.16 ± 0.40^{Ab}	-1.68 ± 0.17^{Cc}	6.29 ± 0.54^{Cc}	7.12 ± 2.59^{Ab}	1.28 ± 0.08^{Ab}
VEO0.25	0.082 ± 0.005^{Aa}	91.30 ± 0.43^{Ab}	-0.86 ± 0.10^{Aa}	3.44 ± 0.06^{Aa}	1.37 ± 0.55^{Aa}	0.84 ± 0.34^{Ba}
VEO0.5	0.090 ± 0.009^{ABa}	91.18 ± 0.56^{Ab}	-0.90 ± 0.08^{ABa}	3.46 ± 0.32^{Aa}	1.65 ± 1.48^{Aa}	1.06 ± 0.12^{Ba}
VEO0.75	0.097 ± 0.019^{Ab}	90.43 ± 0.30^{Abc}	-0.95 ± 0.10^{ABa}	3.70 ± 0.08^{Aa}	3.48 ± 0.78^{Aab}	1.20 ± 0.29^{Aa}
VEO1	0.099 ± 0.010^{Ab}	89.95 ± 0.56^{Ac}	-1.22 ± 0.07^{Bb}	4.36 ± 0.09^{Bb}	4.50 ± 1.48^{Ab}	1.23 ± 0.29^{Aa}

Means in the same column followed by similar lowercase superscripts are not significantly different ($p > 0.05$) between concentrations of EO. Means in the same column followed by similar uppercase superscripts are not significantly different ($p > 0.05$) between types of EO.

were observed visually. It was also found that the L* values were affected by the different types of EOs incorporated to the starch films.

The decrease in L values* might have been due to the increase in diffuse reflectance brought about by light scattering in lipid droplets, particularly EOs, which lowered the light scattering intensity and the films' whiteness index. The study by Šuput *et al.* (2016) also revealed lower L* values at higher concentrations of oregano and black cumin EOs in corn starch films that caused the films to become less transparent and more opaque.

In general, the incorporation of EOs in varying concentrations slightly decreased and increased the values of a* and b*, respectively. The a* values decreased from -0.074 to -0.83, -1.68, and -1.22 when 1% (v/v) of OEO, CEO, and VEO, respectively, were incorporated into the starch films. The lower a* values of the films indicated that the films became slightly greener, especially at a high concentration of EOs. Meanwhile, films incorporated with 1% (v/v) of OEO, CEO, and VEO exhibited b* values of 3.85, 6.29, and 4.36, respectively, which were higher ($p < 0.05$) than that of the control films (3.38), thus indicating an increase in the yellowness of the films. When comparing the different types of EOs, there was a significant difference in a* and b* values at a high concentration of EOs. CEO exhibited the lowest a* and the highest b* values as compared to OEO and VEO, perhaps due to the original colour of oils whereby CEO exists in transparent yellow colour while OEO and VEO exist in almost clear transparent colour when observed visually.

Furthermore, the total colour difference, ΔE, was calculated to distinguish the colour differences between the control starch films and starch/EO films as tabulated in Table 1. The colour of the control films without the incorporation of EOs was used as the reference. As expected, there was an increasing trend of total colour difference with the increase in EOs concentrations incorporated into the films. For starch/OEO, starch/CEO, and starch/VEO films, the total colour difference increased from 1.86 to 6.72, 1.30 to 7.12, and 1.37 to 4.50, respectively, when the concentrations of the EOs were increased from 0.25 to 1% (v/v), consistent with the changes in L*, a*, and b* values. The trend of the ΔE obtained in the present work agreed with the work of Lee *et al.* (2019) which investigated the incorporation of oregano EO nanoemulsion into hydroxypropyl methylcellulose (HPMC) films. Nonetheless, the colour differences were relatively small, and when observed visually, there was almost no discernible change in visual appearance across the film samples.

The opacity of the films is another criterion that has to be considered for food packaging applications which will affect consumers' acceptance. Opacity is related to transparency as high opacity equals low transparency, thus resulting in the product's visibility inside the package. As tabulated in Table 1, the opacity of starch films increased slightly ($p < 0.05$) with the incorporation of EOs, which indicated a decrease in film transparency. Opacity for starch films increased from 0.79 for control films to 1.45, 1.28, and 1.23 when incorporated with 1% (v/v) of OEO, CEO, and VEO, respectively. The opacity of starch/OEO and starch/VEO films were not affected with varying concentrations ($p > 0.05$) of the EOs into the films, but for starch/CEO films, the opacity increased with the increase in EO concentration due to the original colour of CEO as previously mentioned. Nonetheless, the variation was small and unnoticeable. The differences in the opacity might have been due to the coalescence, light scattering, and creaming effect induced by the distribution of lipid molecules particularly EOs in the films, which affected the surface roughness and heterogeneity of the film samples (Akhter *et al.*, 2019). The thickness of the films also contributed to the change in film's opacity where opacity was calculated from the absorbance value at 600 nm per thickness (mm). Starch/OEO films exhibited higher opacity than starch/CEO and starch/VEO films perhaps due to a more compact molecular arrangement within the matrix of the starch/OEO films.

Lee *et al.* (2020) reported similar behaviour with mung bean starch films incorporated with sunflower seed oil. The presence of EO droplets in the matrix of the starch films resulted in blocking of light passage or light scattering, thus increasing the opacity and enhancing the ultraviolet (UV) barrier properties of the films. It is worth noting that the increase in opacity or the decrease in the transparency of the films could be beneficial as it can effectively block UV radiation. This can contribute towards prolonging the shelf life of lipid-rich foods that are vulnerable to the oxidative degradation catalysed by UV radiation.

Another study by Pirouzifard *et al.* (2019), which investigated the incorporation of Zedo gum and *Salvia officinalis* EOs in potato starch films, found an increase in opacity, and a decrease in transparency of films with EOs incorporation. This was due to the formation of a rugged surface on the films during drying. As a result, the EOs accumulated on the surface of the films, and caused heterogeneity in the films.

Film mechanical properties

The effects of different types and concentrations of EOs on the mechanical properties of the films particularly TS, EAB, and YM are presented in Table 2. The tensile strength or TS is defined as the maximum stress that a film can sustain without a break. Table 2 shows that the TS of starch films with the EOs incorporation was significantly lower ($p < 0.05$) than control films. The TS of the films decreased with increasing Eos concentrations, and the decrease became prominent at higher concentrations. The TS decreased from 10.10 MPa for starch/OEO0.25 films to 2.78 MPa for starch film/OEO1 films, from 7.37 MPa for starch/CEO0.25 to 2.88 MPa for starch/CEO1, and from 6.09 MPa for starch/VEO0.25 films to and 1.65 MPa for starch/VEO1 films.

The decreasing value of TS with the incorporation of EOs could have been due to the plasticising effect of EOs, whereby the oil incorporated into the film matrix was responsible for the partial substitution of strong intermolecular polymer-polymer interactions with weak intermolecular polymer-oil interactions, thus resulting in heterogeneity and discontinuities of the films' structure (Nordin *et al.*, 2020). The higher the concentrations of the EOs incorporated into the films, the greater the plasticising effect within the films. The complex structures formed decreased the starch network's cohesion forces which decreased the TS values. The mechanical properties of the films are dependent not only on the type of film-forming material but also on structural cohesion. The oil present in the starch film matrix diminished the cohesion of the polymer network forces, and increased the film matrix's amorphous regions, thus reducing its crystallinity (Lee *et al.*, 2020), and hence reduced the TS.

Similar behaviour was also observed by Šuput *et al.* (2016) who investigated the effects of oregano and black cumin EOs in corn starch films. They revealed that the TS value for the control films was 14.43 MPa, while the TS of films with black cumin EO decreased from 10.02 to 2.3 MPa, and those with oregano EO decreased from 10.65 to 2.12 MPa as the concentrations of EOs were increased from 0.5 to 2%. Furthermore, the decreasing TS values of the films

Table 2. TS, EAB, and YM values of the films.

Film	TS (MPa)	EAB (%)	YM (MPa)
Control	11.87 ± 0.06^{a}	2.34 ± 0.10^{a}	6.03 ± 0.36^{a}
OEO0.25	10.10 ± 1.08^{Ab}	5.61 ± 0.68^{Aa}	4.26 ± 0.75^{Ab}
OEO0.5	3.42 ± 0.24^{Bc}	12.46 ± 2.88^{Ab}	1.64 ± 0.87^{Bc}
OEO0.75	3.13 ± 0.28^{Bc}	18.16 ± 1.24^{Abc}	1.18 ± 0.34^{Cc}
OEO1	2.78 ± 0.04^{Ac}	22.71 ± 4.00^{Ac}	0.80 ± 0.09^{Bc}
CEO0.25	7.37 ± 0.07^{Bb}	2.54 ± 0.12^{Ba}	5.26 ± 0.23^{Aa}
CEO0.5	6.43 ± 0.71^{Ab}	2.91 ± 0.28^{Bab}	4.06 ± 0.50^{Ab}
CEO0.75	4.95 ± 0.11^{Ac}	3.99 ± 0.44^{Cb}	3.84 ± 0.30^{Ab}
CEO1	2.88 ± 0.46^{Ad}	8.58 ± 0.95^{Cc}	2.64 ± 0.07^{Ac}
VEO0.25	6.09 ± 0.15^{Bb}	4.16 ± 0.79^{Ab}	3.03 ± 0.31^{Bb}
VEO0.5	5.17 ± 0.56^{Ab}	10.54 ± 1.10^{Ac}	2.52 ± 0.24^{Bb}
VEO0.75	5.14 ± 0.61^{Ab}	11.70 ± 0.11^{Bc}	2.29 ± 0.35^{Bb}
VEO1	1.65 ± 0.06^{Bc}	16.19 ± 0.07^{Bd}	0.32 ± 0.01^{Cc}

Means in the same column followed by similar lowercase superscripts are not significantly different ($p > 0.05$) between concentrations of EO. Means in the same column followed by similar uppercase superscripts are not significantly different ($p > 0.05$) between types of EO.

were also observed by Al-Hashimi *et al.* (2020) who studied the effects of the incorporation of clove EO into millet starch films, and found that the TS of the millet starch films decreased gradually as clove EO concentrations increased (1 to 3%), ranging from 8.60 to 4.40 MPa.

The results showed that the highest TS decrement was observed for the films with the incorporation of VEO, followed by OEO and then CEO at the concentration of 1% (v/v). Starch/CEO1 films exhibited the highest TS among all the films which could be related to the film's thickness. Starch/CEO1 films exhibited the highest thickness and TS followed by starch/OEO1 films and starch/VEO1 films. The thicker films caused the polymer matrix to become denser and higher in inter- and intramolecular interactions, and consequently became more resistant to rupture (Norfarahin *et al.*, 2018). Moreover, denser films of starch/CEO were evidenced by the high °Brix value as previously mentioned. The CEO contained high total soluble solids, thus making the film thicker and more resistant to rupture.

Meanwhile, the incorporation of EOs into the starch films also affected the EAB values, as shown in Table 2. The EAB is the maximum change in length of the films from their original length before breaking. The EAB value for the control films without the incorporation of EOs was significantly lower ($p < 0.05$) than the films with EOs. The EAB values increased from 2.39% for control films to 22.71, 8.58, and 16.19% for films incorporated with 1% (v/v) OEO, CEO, and VEO, respectively. It can also be seen that there was a trend of increasing EAB with increasing concentrations of EOs incorporated into the starch films.

The increase in EAB indicated that the flexibility of the films was improved with the incorporation of EOs. This could have been due to the presence of oils that play a role as plasticisers or lubricants in the hydrocolloid matrix (Al-Hashimi *et al.*, 2020). The EOs are liquid at room temperature, which is in the form of easily deformed oil droplets in the films, thus increasing the films' extensibility (Song *et al.*, 2018). Moreover, the strong intermolecular polymer-polymer interactions were partially replaced by the weaker polymer-oil interactions, thus generating more flexible domains within the films. The increase in flexibility was also related to structural changes in the starch network because the matrix became less dense, and was placed under tension, thus facilitating the mobility of the polymer chains. The results obtained were consistent with the findings by Song *et al.* (2018) who investigated the incorporation of lemon EO on iron yam and maize starch films, and Lee *et al.* (2019) who investigated the effects of oregano EO incorporation into HPMC films.

EAB value for starch/CEO1 films was lower (8.58%) as compared to starch/OEO1 (22.71%) and starch/VEO1 (16.19%) films, in accordance with the higher TS (2.88 MPa) of the starch/CEO1 films. This indicated that the films exhibited a high strength to fracture but were less flexible. This could have been due to the high molecular weight of the oil compositions in the CEO. The major composition of CEO is known as linalool (32.83% of the oil), which has a high molecular weight of 154.25 g/mol (Rajendran *et al.*, 2014). The high molecular weight is related to larger chains that are entangled, thus giving strength to the films (Balani *et al.*, 2015). Zahiruddin *et al.* (2019) in their work revealed that the high molecular weight plasticiser caused an increase in molecular mobility of the amorphous region, and improved crystallisation of the starch matrix, thus producing strong and rigid films. However, this condition made the film less flexible. On the other hand, the lower molecular weight of one of the major components in OEO, known as carvacrol (14.5% of the oil with a molecular weight of 150.22 g/mol), made the starch films become more flexible than films incorporated with CEO and VEO (28% decanal with a molecular weight of 156.27 g/mol). The cohesive strength of the films decreased when small carvacrol droplets were homogeneously dispersed in the film matrix, thus contributing to the decrease in TS and increase in the films' flexibility.

The incorporation of EOs into the starch films also affected the YM values of the films, as shown in Table 2. YM indicates the stiffness of a material. Generally, it represents how easily the material can bend or stretch. Low YM values indicate that the films exhibit high elasticity (Shapi'i and Othman, 2016). The incorporation of EOs inyo the starch films significantly decreased the YM of the films. The decrease in the YM values became prominent at higher concentrations. The YM values decreased from 6.03 MPa for the control films to 0.80, 2.64, and 0.32 MPa when incorporated with 1% (v/v) of OEO, CEO, and VEO, respectively. There was a decreasing trend of YM when the concentrations of the EOs incorporated into the starch films were increased. The decrease in YM values demonstrated that the incorporation of EOs to the films made the films become less stiff, and exhibited improvement in flexibility. The higher the concentrations of EOs incorporated into the films, the less stiff and the more elastic the films became.

The trend of YM values obtained in the present work was similar to that of Lee *et al.* (2019) who investigated the effects of oregano EO incorporation into HPMC films. The incorporation of oregano EOs within the HPMC biopolymer matrices decreased the YM of the films due to the structural deformity within the biopolymer matrices, which consequently decreased the fracture strength of the films. Apart from that, EOs acted as plasticisers, increasing the mobility of starch chains, thus reducing film's rigidity and stiffness (Nordin *et al.*, 2020). Basiak *et al.* (2016) also found a decrease in YM values for starch-based films laminated with rapeseed oil.

The measured YM values exhibited a similar trend to that of TS, whereby starch/CEO1 films were stiffer than starch/OEO1 and starch/VEO1 films. While the TS measures how much force or stress a material can withstand before failure, YM measures how the strength changes as a material is stretched. As earlier stated, starch/CEO1 films were denser and thicker due to the oil compositions of the CEO, which resulted in films that had high resistance to fracture. This condition made the starch/CEO1 films exhibit a high modulus and stiffer characteristics. Incorporating VEO at 1% (v/v) into the starch films produced films with the lowest YM value, thus indicating that the films were less stiff and more flexible than other starch/EO films. The YM is the ratio of tensile stress and strain; hence, the YM decreased as the strain increased. Starch/VEO1 films had the lowest YM due to the highest value of strain or EAB (16.19%).

Quality attributes of strawberries

The potential application of starch/EOs films as food packaging materials was demonstrated on strawberries, stored in a chiller at 4°C for 9 d. The quality attributes of the strawberries were investigated in terms of weight loss, colour changes, and firmness.

Weight loss

The weight loss of strawberries is a factor that will affect the quality and impact consumers' preferences. Weight loss may affect the quality changes of strawberries in terms of colour and texture. The percentage weight losses of the wrapped and unwrapped strawberries are shown in Table 3.

Table 3. Percentage of weight loss (%) of strawberries during 9-day storage.

Sample	Day 3 (%)	Day 6 (%)	Day 9 (%)
Unwrapped	26.71 ± 0.02^{A}	35.15 ± 0.20^{B}	42.44 ± 0.01^{C}
Control	17.16 ± 0.14^{Aa}	21.69 ± 0.05^{Ba}	24.79 ± 0.10^{Ca}
OEO0.25	14.31 ± 0.17^{Aba}	19.70 ± 0.09^{Bba}	21.54 ± 0.10^{Cba}
OEO0.5	12.87 ± 0.06^{Aca}	18.22 ± 0.13^{Bca}	20.66 ± 0.06^{Cca}
OEO0.75	10.08 ± 0.12^{Ada}	17.86 ± 0.05^{Bcda}	19.61 ± 0.02^{Cda}
OEO1	9.69 ± 0.03^{Aea}	17.53 ± 0.41^{Bda}	19.17 ± 0.12^{Cda}
CEO0.25	16.10 ± 0.02^{Abb}	18.90 ± 0.04^{Bba}	21.49 ± 0.10^{Cda}
CEO0.5	14.66 ± 0.06^{Acb}	17.83 ± 0.08^{Bca}	20.60 ± 0.01^{Cca}
CEO0.75	13.80 ± 0.05^{Adb}	17.25 ± 0.10^{Bda}	19.69 ± 0.04^{Cda}
CEO1	11.22 ± 0.09^{Aeb}	15.25 ± 0.01^{Beb}	18.20 ± 0.01^{Cea}
VEO0.25	16.83 ± 0.71^{Aab}	20.38 ± 0.01^{Bba}	22.91 ± 0.05^{Cba}
VEO0.5	16.51 ± 0.05^{Aabc}	19.40 ± 0.05^{Bca}	20.49 ± 0.04^{Cca}
VEO0.75	15.56 ± 0.11^{Abb}	18.22 ± 0.03^{Bdb}	19.54 ± 0.01^{Cda}
VEO1	10.13 ± 0.11^{Acb}	14.78 ± 0.09^{Beb}	18.68 ± 0.12^{Cea}

Means in the same column followed by similar lowercase italic superscripts are not significantly different ($p > 0.05$) between concentrations of EOs. Means in the same column followed by similar lowercase superscripts are not significantly different ($p > 0.05$) between types of EOs. Means in the same row followed by similar uppercase superscripts are not significantly different ($p > 0.05$) between storage days.

As expected, there was a gradual increase in weight loss of the strawberries with storage time. However, during the storage period, the percentage weight loss for the unwrapped strawberries was more pronounced than for strawberries wrapped with starch (control) and starch/EOs films. In general, the percentage weight loss of strawberries wrapped with starch/EO films was lower than the strawberries wrapped with control films. The lowest percentage weight loss was found for strawberries wrapped in starch/EOs films incorporated with 1% (v/v) EOs throughout 9-day storage. For example, the percentage weight losses for strawberries wrapped with films incorporated with 1% (v/v) OEO, CEO, and VEO were 19.17, 18.20, and 18.68%, respectively, at day 9 of storage. These results were lower than that of the strawberries wrapped with the control films and unwrapped strawberries which were 24.79 and 42.44%, respectively. It can also be seen from Table 3 that the percentage weight loss of the strawberries decreased with increasing concentrations of EOs incorporated into the starch films. For instance, on day 9, the percentage weight losses of strawberries were 21.54, 20.66, 19.61, and 19.17% when wrapped with films incorporated with 0.25, 0.5, 0.75, and 1% (v/v) of OEO, respectively.

The increase in physiological weight loss in fruits, including strawberries, was primarily due to the moisture loss caused by the respiration and transpiration processes throughout storage (Abbasi *et al.*, 2009). Aziman *et al.* (2021) highlighted that weight loss in packaged food could also be associated with enzymatic reaction and microbial activity occurring during storage, thus influencing the weight changes. The lower percentage in weight loss of strawberries wrapped in the films as compared to the unwrapped strawberries indicated that the films played an important role as a water vapour barrier between the strawberries and the environment throughout the storage period (Cai *et al.*, 2020). Without the film's barrier, it would be easy for the water in strawberries to be lost to the surrounding. The existence of the film's barrier was also found to be able to prevent water transfer and protect the strawberries' skin from mechanical injury, thus resulting in delaying water loss (Dhital *et al.*, 2018). The unwrapped strawberries encountered an acceleration of weight loss which can be attributed to the increase in the metabolic activity associated with the desiccation of fruit cells during long storage (Ali *et al.*, 2014) which slowed down following wrapping (Sanchez-Gonzalez *et al*., 2011).

Meanwhile, the percentage weight loss of strawberries wrapped with starch/EOs films was lower than strawberries wrapped with the starch films without EOs. This could have been due to the lower water vapour permeability (WVP) of starch films incorporated with EOs than those without EOs incorporated. Pelissari *et al.* (2009) found that cassava starch films exhibited a high WVP value of 1.39×10^{-10} g/Pa.m.s, but the WVP value decreased significantly with increasing OEO concentrations in the films whereby the WVP of cassava starch films for maximum concentration of OEO (1%, w/w) was 0.62×10^{-10} g/Pa.m.s, thus supporting the results observed in the present work. Apart from that, Ibrahim *et al.* (2017) demonstrated that the coatings of strawberries with chitosan films incorporated with lemongrass and thyme EOs at the concentrations of 0.1 and 0.2% (v/v) decreased the weight loss of strawberries. This could have been due to the high WVP barrier properties associated with the incorporation of EOs in the coatings.

In the present work, the percentage weight loss was the least for the strawberries wrapped with starch/CEO1 films followed by that of starch/VEO1, and then by starch/OEO1 films. Additionally, the thickness of the films might have contributed to the decrease in percentage weight loss of the wrapped strawberries. The thicker starch/CEO1 films might have exhibited high barrier properties and protected the strawberries from mechanical damage, which decreased the water loss.

Colour changes

The colour of the food product, in this case, strawberry, is an important attribute for the consumers' acceptance. The red colour in strawberries is associated with anthocyanin accumulation in ripe fruits (Dhital *et al.*, 2018). In the present work, the colour of the strawberries was evaluated by recording the L* and a* values, as tabulated in Table 4. In general, the L* and a* values decreased with the progression of storage period for all the strawberries. The percentage decrease of L* and a* values of strawberries after 9-day storage was more pronounced for unwrapped strawberries (36.99 and 50.86%, respectively) as compared to all the wrapped strawberries.

The percentage decrease of L* values for strawberries wrapped with starch films (34.77%) was higher than strawberries wrapped with starch/EOs films, whereby the percentage decrease for strawberries wrapped with starch/EOs films were 26.1, 26.85, and 19.98% for starch/OEO1, starch/CEO1, and starch/VEO1 films, respectively. The L* values of the strawberries wrapped with starch/EOs films were also higher than that of control films. For instance, on day 9, the L* values of strawberries wrapped with starch films incorporated with 1% (v/v) OEO, CEO, and VEO were 34.73, 35.16, and 34.60, respectively, as compared to 34.16 for strawberries wrapped with control films. This trend was more significant for films containing higher concentrations of EOs, thus proving the starch/EOs films' effectiveness in preventing colour deterioration of the strawberries by decreasing the anthocyanin accumulation, and delaying the senescence of fruits (Shehata *et al.*, 2020). Also, the changes in the L* values of the strawberries are indicative of oxidative reaction. The oxidative reaction induces enzymatic and non-enzymatic browning effects, thus leading to the loss of the fruit's natural colour (Chansiw *et al.*, 2018). It can be deduced that the starch/EO films were capable of decreasing the oxidative reaction due to the higher oxygen barrier properties as compared to starch films without EO.

When comparing the different types of starch/EOs films, starch/VEO films were the most effective films especially at a high concentration (1%, v/v) as the strawberries wrapped with the films had the lowest decrease in L* values during 9-day storage. This might have been due to the presence of antioxidant compounds in the VEO that had the highest antioxidant activity (52.59 mg/g extract) which could delay the colour deterioration of the strawberries as compared to that of OEO (49.1 mg/g extract) and CEO (17.52 mg/g extract) (Chansiw *et al.*, 2018).

Meanwhile, the decrease in a* values was pronounced for unwrapped strawberries, whereby the a* values decreased from 23.28 to 8.58 (50.86%), followed by wrapped strawberries in control films where the a* values decreased from 20.26 to 10.45 (48.42%) throughout 9-day storage. The lowest percentage decrease in a* values after 9-day storage was found for strawberries wrapped with starch/EOs films whereby the a* values decreased from 18.90 to

Table 4. L* and a* values of strawberries during 9-day storage.

Sample	Day 0	Day 3	Day 6	Day 9
L* values				
Unwrapped	$43.24 \pm 0.47^{\mathrm{A}}$	$37.92 \pm 1.79^{\mathrm{B}}$	$34.35 \pm 1.15^{\mathrm{C}}$	$27.24 \pm 1.33^{\mathrm{D}}$
Control	$48.23 \pm 0.83^{\mathrm{A}a\mathrm{a}}$	$38.45 \pm 1.24^{\mathrm{B}a\mathrm{a}}$	$35.14 \pm 1.17^{\mathrm{B}a}$	$31.46 \pm 0.43^{\mathrm{C}a\mathrm{a}}$
OEO0.25	$47.00 \pm 2.46^{\mathrm{A}a\mathrm{a}}$	$39.06 \pm 0.40^{\mathrm{B}b\mathrm{a}}$	$36.21 \pm 0.46^{\mathrm{B}ab\mathrm{a}}$	$31.69 \pm 1.69^{\mathrm{C}a\mathrm{a}}$
OEO0.5	$47.98 \pm 0.15^{\mathrm{A}a\mathrm{a}}$	$39.82 \pm 0.55^{\mathrm{B}b\mathrm{a}}$	$36.78 \pm 0.47^{\mathrm{B}ab\mathrm{a}}$	$31.70 \pm 0.47^{\mathrm{C}a\mathrm{a}}$
OEO0.75	$48.52 \pm 0.31^{\mathrm{A}a\mathrm{a}}$	$39.99 \pm 0.57^{\mathrm{B}b\mathrm{a}}$	$37.48 \pm 0.92^{\mathrm{B}b\mathrm{b}}$	$34.36 \pm 3.16^{\mathrm{C}a\mathrm{b}}$
OEO1	$47.00 \pm 0.50^{\mathrm{A}a\mathrm{a}}$	$40.57 \pm 1.18^{\mathrm{B}b\mathrm{b}}$	$37.95 \pm 0.32^{\mathrm{B}b\mathrm{b}}$	$34.73 \pm 0.69^{\mathrm{C}b\mathrm{a}}$
CEO0.25	$47.78 \pm 1.25^{\mathrm{A}a\mathrm{a}}$	$39.45 \pm 0.90^{\mathrm{B}ab\mathrm{a}}$	$36.09 \pm 0.79^{\mathrm{B}ab\mathrm{a}}$	$31.57 \pm 0.19^{\mathrm{C}ab\mathrm{a}}$
CEO0.5	$47.620 \pm 1.00^{\mathrm{A}a\mathrm{a}}$	$39.63 \pm 1.96^{\mathrm{B}ab\mathrm{a}}$	$37.04 \pm 1.10^{\mathrm{B}ab\mathrm{a}}$	$31.82 \pm 2.17^{\mathrm{C}ab\mathrm{a}}$
CEO0.75	$47.55 \pm 1.38^{\mathrm{A}a\mathrm{a}}$	$40.45 \pm 0.42^{\mathrm{B}ab\mathrm{a}}$	$37.47 \pm 1.08^{\mathrm{B}bc\mathrm{a}}$	$32.47 \pm 2.37^{\mathrm{C}ab\mathrm{a}}$
CEO1	$48.06 \pm 0.67^{\mathrm{A}a\mathrm{a}}$	$42.35 \pm 2.81^{\mathrm{B}b\mathrm{a}}$	$39.06 \pm 0.44^{\mathrm{B}c\mathrm{a}}$	$35.16 \pm 2.86^{\mathrm{C}b\mathrm{a}}$
VEO0.25	$47.96 \pm 1.35^{\mathrm{A}a\mathrm{a}}$	$39.70 \pm 0.37^{\mathrm{B}ab\mathrm{b}}$	$35.26 \pm 0.20^{\mathrm{C}a\mathrm{a}}$	$32.12 \pm 0.64^{\mathrm{D}b\mathrm{a}}$
VEO0.5	$46.87 \pm 0.87^{\mathrm{A}ab\mathrm{a}}$	$40.23 \pm 0.64^{\mathrm{B}ab\mathrm{a}}$	$35.34 \pm 0.47^{\mathrm{B}a\mathrm{a}}$	$33.06 \pm 0.67^{\mathrm{B}b\mathrm{a}}$
VEO0.75	$45.47 \pm 0.57^{\mathrm{A}b\mathrm{b}}$	$41.01 \pm 0.98^{\mathrm{B}ab\mathrm{a}}$	$36.31 \pm 0.24^{\mathrm{C}a\mathrm{a}}$	$34.91 \pm 0.58^{\mathrm{C}c\mathrm{b}}$
VEO1	$43.24 \pm 0.36^{\mathrm{A}c\mathrm{b}}$	$42.05 \pm 3.68^{\mathrm{B}b\mathrm{a}}$	$36.48 \pm 1.46^{\mathrm{C}a\mathrm{b}}$	$34.6 \pm 0.19^{\mathrm{C}c\mathrm{a}}$
a* values				
Unwrapped	$23.28 \pm 2.94^{\mathrm{A}a}$	$15.62 \pm 1.63^{\mathrm{A}}$	$11.20 \pm 0.63^{\mathrm{B}}$	$8.58 \pm 0.50^{\mathrm{B}}$
Control	$20.26 \pm 1.31^{\mathrm{A}}$	$15.92 \pm 0.79^{\mathrm{B}a}$	$13.20 \pm 0.63^{\mathrm{B}a}$	$10.45 \pm 0.56^{\mathrm{C}a}$
OEO0.25	$21.52 \pm 0.50^{\mathrm{A}a\mathrm{a}}$	$16.78 \pm 0.85^{\mathrm{B}ab\mathrm{a}}$	$13.41 \pm 2.08^{\mathrm{C}a\mathrm{b}}$	$11.87 \pm 0.66^{\mathrm{C}b\mathrm{a}}$
OEO0.5	$21.07 \pm 0.71^{\mathrm{A}a\mathrm{b}}$	$16.84 \pm 3.16^{\mathrm{B}bc\mathrm{a}}$	$13.94 \pm 0.18^{\mathrm{B}ab\mathrm{b}}$	$10.86 \pm 0.96^{\mathrm{C}a\mathrm{a}}$
OEO0.75	$20.21 \pm 0.97^{\mathrm{A}a\mathrm{b}}$	$17.18 \pm 1.85^{\mathrm{B}bc\mathrm{a}}$	$14.38 \pm 1.60^{\mathrm{B}ab\mathrm{b}}$	$11.69 \pm 0.33^{\mathrm{C}b\mathrm{a}}$
OEO1	$18.90 \pm 1.32^{\mathrm{A}a\mathrm{b}}$	$17.77 \pm 0.87^{\mathrm{B}c\mathrm{b}}$	$15.06 \pm 2.02^{\mathrm{B}cb\mathrm{b}}$	$12.32 \pm 1.17^{\mathrm{C}b\mathrm{a}}$
CEO0.25	$19.26 \pm 1.31^{\mathrm{A}ab\mathrm{a}}$	$16.61 \pm 1.32^{\mathrm{B}a\mathrm{a}}$	$18.06 \pm 1.64^{\mathrm{B}b\mathrm{a}}$	$10.89 \pm 0.28^{\mathrm{C}a\mathrm{a}}$
CEO0.5	$17.84 \pm 0.79^{\mathrm{A}b\mathrm{a}}$	$16.62 \pm 1.63^{\mathrm{A}a\mathrm{a}}$	$18.92 \pm 0.79^{\mathrm{B}b\mathrm{a}}$	$11.43 \pm 0.50^{\mathrm{B}ab\mathrm{a}}$
CEO0.75	$12.67 \pm 1.25^{\mathrm{A}c\mathrm{a}}$	$18.67 \pm 1.25^{\mathrm{B}b\mathrm{a}}$	$19.86 \pm 0.32^{\mathrm{B}c\mathrm{a}}$	$11.03 \pm 0.95^{\mathrm{A}ab\mathrm{a}}$
CEO1	$18.93 \pm 0.63^{\mathrm{A}c\mathrm{a}}$	$19.86 \pm 0.32^{\mathrm{AB}b\mathrm{a}}$	$19.88 \pm 1.07^{\mathrm{B}c\mathrm{a}}$	$11.86 \pm 0.78^{\mathrm{B}b\mathrm{a}}$
VEO0.25	$21.05 \pm 1.52^{\mathrm{A}a\mathrm{a}}$	$15.46 \pm 2.61^{\mathrm{AB}a\mathrm{a}}$	$13.22 \pm 0.77^{\mathrm{B}a\mathrm{b}}$	$10.54 \pm 0.67^{\mathrm{C}a\mathrm{a}}$
VEO0.5	$19.83 \pm 0.23^{\mathrm{A}ab\mathrm{ab}}$	$16.24 \pm 0.30^{\mathrm{B}a\mathrm{a}}$	$13.89 \pm 0.70^{\mathrm{B}a\mathrm{b}}$	$12.62 \pm 1.09^{\mathrm{C}b\mathrm{a}}$
VEO0.75	$18.50 \pm 2.69^{\mathrm{A}ab\mathrm{b}}$	$17.59 \pm 0.68^{\mathrm{B}b\mathrm{a}}$	$14.38 \pm 1.27^{\mathrm{B}ab\mathrm{b}}$	$12.32 \pm 1.32^{\mathrm{C}b\mathrm{a}}$
VEO1	$17.47 \pm 0.73^{\mathrm{A}b\mathrm{b}}$	$19.75 \pm 1.27^{\mathrm{A}c\mathrm{a}}$	$15.28 \pm 0.36^{\mathrm{B}b\mathrm{b}}$	$13.59 \pm 0.67^{\mathrm{B}b\mathrm{b}}$

Means in the same column followed by similar lowercase italic superscripts are not significantly different ($p > 0.05$) between concentrations of EOs. Means in the same column followed by similar lowercase superscripts are not significantly different ($p > 0.05$) between types of EOs. Means in the same row followed by similar uppercase superscripts are not significantly different ($p > 0.05$) between storage days.

12.32 (34.81%), from 18.93 to 11.86 (37.35%), and from 17.47 to 13.59 (22.20%) for starch/OEO1, starch CEO1, and starch VEO1 films, respectively. The decreasing trend of a* values demonstrated that the strawberries lost their redness during the storage period due to the factors such as loss of red anthocyanin pigment and the formation of brown pigments (Abers and Wrolstad, 1979). The strawberries wrapped with starch/EOs films exhibited less decrease in a* values and high a* values at the end of storage, thus indicating less decrease in the fruit's redness because the films were able to prevent the oxidation and browning enzymatic reactions due to the presence of antioxidants components in the EOs. Ali *et al.* (2015) were in agreement, whereby they observed that the application of propolis coating was able to preserve the colour of bell pepper, which is indicative of the fruit's slow ripening physiology under storage conditions. When comparing different types of starch/EOs films, the results showed that starch films with the incorporation of VEO at 1% (v/v) concentration were the most effective in controlling the redness deterioration of the strawberries, consistent with the findings related to L* values.

Figure 1 shows the changes in appearance of wrapped and unwrapped strawberries on days 0, 3, 6, and 9 of storage in a chiller at 4°C. The appearance seemed to be consistent with the discussion on L* and a* values. The strawberries became slightly dark in colour at the end of storage. The red colour of the unwrapped strawberries were darker than the wrapped strawberries, and mould started to grow on day 9 of storage. At the end of storage, the red colour of strawberries wrapped in control films were slightly darker than starch/EO films. When comparing among the different types of starch/EOs films, it was observed that strawberries wrapped with starch/VEO1 films were more effective as compared to others at the end of storage, as the strawberries exhibited the highest lightness, and the redness of the strawberries was maintained.

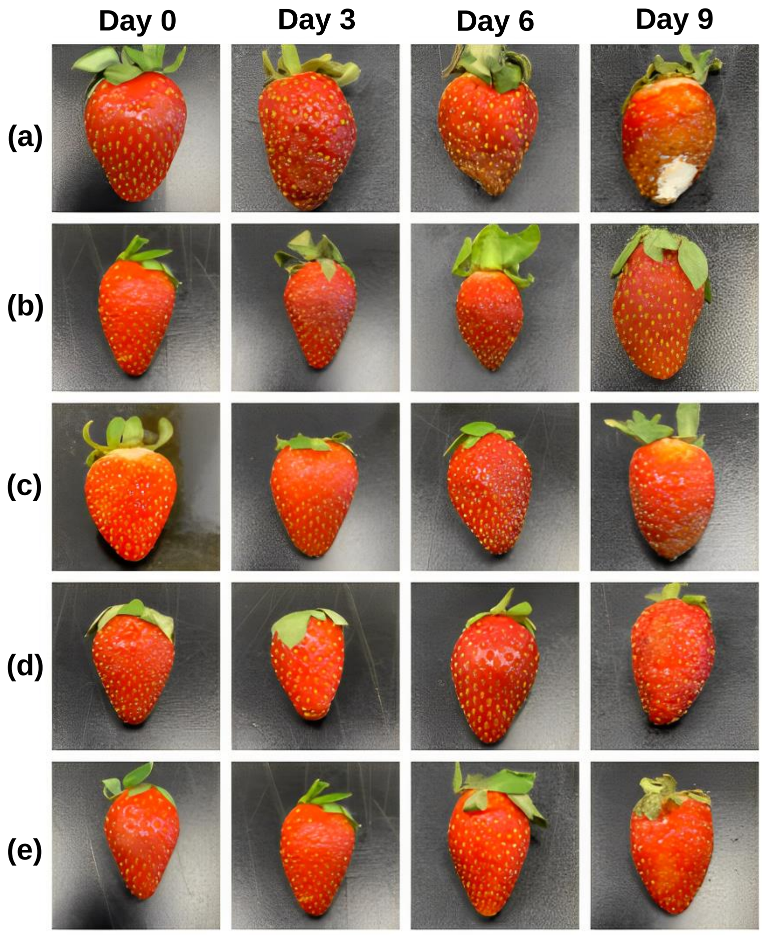

Figure 1. Appearance changes of strawberries on days 0, 3, 6, and 9 of storage at 4°C with **(a)** unwrapped, **(b)** wrapped with starch films, **(c)** wrapped with starch/OEO1 films, **(d)** wrapped with starch/CEO1 films, and **(e)** wrapped with starch/VEO1 film.

Firmness

Fruit firmness is another important quality attribute of strawberries that will contribute to consumers' acceptance. The loss of firmness is one of the main factors which limits the quality and postharvest shelf life of strawberries. Strawberries soften during ripening by degradation of the middle lamella of the cell wall of cortical parenchyma cells (Niu *et al.*, 2019). Table 5 tabulates the firmness in Newton (N) of the strawberries throughout 9-day storage.

Table 5. Firmness (N) of strawberries during 9-day storage.

Sample	**Day 0 (N)**	**Day 3 (N)**	**Day 6 (N)**	**Day 9 (N)**
Unwrapped	$94.20 \pm 4.43^{\mathrm{A}}$	$77.27 \pm 6.53^{\mathrm{B}}$	$48.31 \pm 2.47^{\mathrm{C}}$	$36.46 \pm 3.15^{\mathrm{D}}$
Control	$76.30 \pm 3.43^{\mathrm{A}a}$	$58.04 \pm 9.79^{\mathrm{AB}a}$	$48.62 \pm 9.77^{\mathrm{B}a}$	$43.60 \pm 3.07^{\mathrm{B}a}$
OEO0.25	$77.77 \pm 2.53^{\mathrm{A}ab\mathrm{a}}$	$66.78 \pm 1.63^{\mathrm{B}a\mathrm{a}}$	$55.46 \pm 2.81^{\mathrm{C}ab\mathrm{b}}$	$45.36 \pm 3.79^{\mathrm{D}a\mathrm{b}}$
OEO0.5	$84.02 \pm 4.00^{\mathrm{A}ab\mathrm{b}}$	$69.18 \pm 5.16^{\mathrm{AB}a\mathrm{a}}$	$64.00 \pm 2.81^{\mathrm{BC}ab\mathrm{b}}$	$50.95 \pm 3.52^{\mathrm{C}ab\mathrm{b}}$
OEO0.75	$84.51 \pm 6.89^{\mathrm{A}ab\mathrm{a}}$	$71.35 \pm 7.51^{\mathrm{AB}a\mathrm{b}}$	$67.62 \pm 4.68^{\mathrm{B}b\mathrm{b}}$	$53.61 \pm 5.68^{\mathrm{C}ab\mathrm{b}}$
OEO1	$89.50 \pm 5.40^{\mathrm{A}b\mathrm{a}}$	$73.26 \pm 4.27^{\mathrm{B}a\mathrm{b}}$	$70.21 \pm 1.05^{\mathrm{B}b\mathrm{b}}$	$59.03 \pm 3.17^{\mathrm{C}b\mathrm{a}}$
CEO0.25	$74.34 \pm 1.42^{\mathrm{A}c\mathrm{a}}$	$64.39 \pm 2.42^{\mathrm{B}ab\mathrm{a}}$	$52.41 \pm 7.42^{\mathrm{C}a\mathrm{a}}$	$48.23 \pm 2.28^{\mathrm{C}ab\mathrm{a}}$
CEO0.5	$74.16 \pm 3.60^{\mathrm{A}c\mathrm{a}}$	$66.94 \pm 4.10^{\mathrm{B}ab\mathrm{a}}$	$57.21 \pm 2.11^{\mathrm{C}a\mathrm{a}}$	$53.53 \pm 0.48^{\mathrm{C}ab\mathrm{a}}$
CEO0.75	$87.05 \pm 3.65^{\mathrm{A}ab\mathrm{a}}$	$75.24 \pm 4.26^{\mathrm{B}b\mathrm{a}}$	$63.77 \pm 3.60^{\mathrm{BC}a\mathrm{a}}$	$55.79 \pm 2.95^{\mathrm{C}b\mathrm{a}}$
CEO1	$88.74 \pm 7.20^{\mathrm{A}a\mathrm{a}}$	$77.25 \pm 5.72^{\mathrm{B}b\mathrm{a}}$	$64.53 \pm 9.35^{\mathrm{BC}a\mathrm{a}}$	$56.65 \pm 8.64^{\mathrm{C}b\mathrm{a}}$
VEO0.25	$92.72 \pm 5.48^{\mathrm{A}b\mathrm{b}}$	$74.56 \pm 4.63^{\mathrm{B}ab\mathrm{b}}$	$57.04 \pm 5.81^{\mathrm{C}a\mathrm{b}}$	$49.22 \pm 8.88^{\mathrm{C}a\mathrm{a}}$
VEO0.5	$116.97 \pm 3.38^{\mathrm{A}c\mathrm{b}}$	$76.41 \pm 4.31^{\mathrm{B}bc\mathrm{b}}$	$59.91 \pm 4.55^{\mathrm{C}a\mathrm{a}}$	$51.47 \pm 3.09^{\mathrm{C}a\mathrm{b}}$
VEO0.75	$108.30 \pm 4.63^{\mathrm{A}c\mathrm{b}}$	$90.30 \pm 4.75^{\mathrm{B}bc\mathrm{b}}$	$79.42 \pm 6.98^{\mathrm{B}b\mathrm{c}}$	$65.30 \pm 4.16^{\mathrm{C}b\mathrm{c}}$
VEO1	$110.22 \pm 9.31^{\mathrm{A}c\mathrm{b}}$	$95.44 \pm 5.04^{\mathrm{B}c\mathrm{c}}$	$86.17 \pm 5.04^{\mathrm{BC}b\mathrm{c}}$	$74.62 \pm 1.51^{\mathrm{C}b\mathrm{b}}$

Means in the same column followed by similar lowercase italic superscripts are not significantly different ($p > 0.05$) between concentrations of EOs. Means in the same column followed by similar lowercase superscripts are not significantly different ($p > 0.05$) between types of EOs. Means in the same row followed by similar uppercase superscripts are not significantly different ($p > 0.05$) between storage days.

As expected, all the strawberries experienced a loss in firmness during the storage period, but the loss in firmness was much higher for unwrapped strawberries whereby the firmness decreased from 94.20 to 36.46 N (61.30%) after 9-day storage. The strawberries wrapped with control films exhibited a lower percentage decrease in firmness as compared to unwrapped strawberries whereby the firmness decreased from 76.30 to 43.60 N (42.85%) after 9-day storage. Meanwhile, the strawberries wrapped with starch/EOs films exhibited the lowest percentage decrease in firmness, whereby the firmness decreased from 89.50 to 59.03N (34.04%) for starch/OEO1 films, from 88.74 to 56.65 N (36.16%) for starch/CEO1 films, and from 110.22 to 74.62 N (32.3%) for starch/VEO1 films. At the end of storage, the firmness for strawberries wrapped with starch/OEO1, starch/CEO1, and starch/VEO1 films was 59.03, 56.65, and 74.62 N, respectively, which were significantly higher than the strawberries wrapped with control films (43.60 N) and unwrapped strawberries (36.46 N). Furthermore, when varying the concentration of EOs, there was a decreasing pattern of the percentage decrease in firmness of the wrapped strawberries with increasing EOs concentrations from 0.25 to 1% (v/v).

This indicated that incorporating EOs in the films could decrease changes in the quality attributes, specifically the firmness of strawberries. Furthermore, the starch films incorporated with the highest concentration of EOs were found to be the most efficient in decreasing the firmness loss of the strawberries. This could have been due to the good barrier properties of the films that decreased the respiration and water loss, thus maintaining firmness. This was supported by Maqbool *et al.* (2013) who demonstrated the efficiency of a 10% gum Arabic and 0.75% chitosan coating in preserving firmness and delaying banana ripening.

The findings observed in the present work were consistent with the study by Yusof *et al.* (2018) in which they investigated the effects of starch-chitosan with turmeric oil coatings on the firmness of strawberries, and found that the coatings managed to control the firmness reduction throughout 7-day storage because the coatings lessened the cell's deterioration during storage time, thus lowering the respiration and other ripening processes. When comparing different types of EOs, starch films with the incorporation of VEO at 1% (v/v) concentration resulted in the lowest decrease in firmness at the end of storage. This might have been due to the antioxidant activity of the EO which prevented the oxidation reaction in the fruits that might cause the loss of firmness, as previously discussed.

Conclusion

The starch/EO films incorporated with different types and concentrations of EOs were produced *via* the solvent casting method. The incorporation of EOs in starch films decreased the lightness, and increased the yellowness and opacity of the films, especially at higher concentrations of EOs. TS and YM of the starch films decreased but the EAB increased with the incorporation of EOs at a high concentration (1%, v/v), revealing that EOs in starch films were able to decrease the film's strength and stiffness, and increased flexibility and elasticity due to the plasticising effect. The starch/CEO1 films exhibited the highest TS and YM due to the thicker and denser films, which were more resistant to fracture. On the other hand, starch/OEO1 films exhibited the highest EAB value due to the high composition of low molecular weight components (carvacrol) in OEO, thus making the films flexible. In terms of the potential application of the films, the incorporation of EOs especially at a high concentration managed to reduce the changes in quality attributes of strawberries, particularly weight loss, colour changes, and the loss of firmness of strawberries throughout 9-day storage. Among the starch/EOs films, the films incorporated with VEO were the most effective because the films exhibited relatively high flexibility and low stiffness apart from exhibiting low opacity. The strawberries wrapped with starch/VEO films exhibited the least decrement in colour, weight loss, and firmness throughout 9-day storage. The incorporation of EOs in the starch films produced packaging materials that had the potential for food packaging applications to extend the shelf life, and maintain the quality attributes of fruits.

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