

Controlled Release of Orange Oil Vapour to Delay the Ripening and Mould Growth of Mangosteens (*Garcinia mangostana*) using Rubberwood Sawdust

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The effect of rubberwood sawdust containing orange oil was evaluated relative to the quality of mangosteens after harvest, transportation, and storage. Orange oil emulsion (500 $\mu\text{L L}^{-1}$) was added to the rubberwood sawdust as an adsorber and inserted into sachets in different amounts (0 g, 70 g, 140 g, 210 g, and 280 g). Then, each sachet was packed into a corrugated paper box (about 35 L) containing 15 kg of mangosteens, transported for 3 days by truck, and stored at 30 ± 2 °C for nine days. Results showed that the sachet containing 210 g of orange oil absorbent was able to delay the ripening of mangosteens by two stages of the ripening scale and inhibit mould growth. In addition, the overall quality of the treated fruit was good, especially in terms of loss of weight and firmness. Orange oil vapour from the sachet had a high antioxidant capacity that affected the pigment of mangosteen by preventing the degradation of chlorophyll and carotenoids. D-limonene was the main factor that inhibited mycelium growth on the fruit. This finding indicates that rubberwood sawdust containing orange oil in a paper box can be used to improve the quality of mangosteens in the fruit market.

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INTRODUCTION

Mangosteen (*Garcinia mangostana*), which is known as the queen of fruits and belongs to the family of Guttiferae, is now widely cultivated in tropical areas such as Indonesia, Malaysia, Sri Lanka, the Philippines, and Thailand (Pedraza-Chaverri *et al.* 2008). It is known that mangosteen is a seasonal fruit that is highly perishable; however, it provides several bioactive compounds (Gorinstein *et al.* 2011), such as provitamin A carotenoids, antioxidant activity, phenolic compounds, dietary fibre, carbohydrates, and minerals for human health (Aizat *et al.* 2019). Mangosteen is a climacteric respiration fruit: when the fruit goes through the climacteric stage, the ripening process occurs with the increasing rate of respiration, the degradation of chlorophyll, and the increase in anthocyanins and change the fruit's skin colour. The ripening process of mangosteen occurs at such a high rate that the peel and the calyxes change rapidly, turning from green to purple within 4 to 7 days of being harvested. Therefore, logistics management is difficult because of the perishability and fragility of mangosteens, and the quality deteriorates quickly before delivery to consumers.

Further, one of the significant post-harvest problems related to mangosteen is that it is easily susceptible to and is attacked by moulds such as *Pestalotiopsis* sp., *Lasiodiplodia theobromae*, *Phomopsis* sp., and *Colletotrichum gloeosporioides* (Khewkhom *et al.* 2013). Contamination of mould occurs in mangosteens during plantation and post-harvest, and it can cause the fruit to be spoiled after harvest within 6 to 9 days (Lerslerwong *et al.* 2013).

Accordingly, serious efforts have been made to prolong the shelf life of mangosteens and reduce fruit wastage and loss during transportation and storage before they reach the consumer. The development of active packaging for transportation and storage by adding essential oils *via* sachets or coating is one of the alternative technologies for fruit preservation to maintain the quality of the fruit (Owolabi *et al.* 2021a; Phothisuwan *et al.* 2021; Saengwong-ngam *et al.* 2022). Orange oil is one of the flavouring ingredients or essential oils for food additives, evaluated by expert panels to determine whether they are “generally recognised as safe” (GRAS) and accepted and used worldwide in the food industry. Orange oil can be extracted from citrus peels as an agro-industry waste material. It is a rich source of bioactive compounds (Li *et al.* 2021). The function of orange oil is to add flavour additives to many food products. It also plays a vital role regarding antioxidant activity (Radünz *et al.* 2021) and the prevention of fungi, such as *Penicillium digitatum* and *Penicillium italicum* (Caccioni *et al.* 1998). However, adding essential oil in packaging to control fruit quality is only applicable to short-term storage due to the quick vaporisation of the essential oils. There is a need to control the vapour phase release process of the essential oils for long-term use.

Rubberwood sawdust, an organic porous adsorbent (Mazlan *et al.* 2016), was selected to adsorb the liquid orange oil and packed in sachets then to gradually release the vapour phase during the transportation and storage of mangosteens. Rubberwood sawdust can efficiently adsorb essential oil. Additionally, its high adsorption capacity, reusability, and low cost make it suitable for large-scale applications (Parichanon *et al.* 2021). It is necessary to assess the effects of ripening regulators together with techniques that regulate the control of mould growth during transportation and storage of the mangosteens. To date, there have been no reports on the influence of orange oil vapour on the ripening colour change of mangosteens. This research aims to develop an essential oil-treated rubberwood sawdust capable of a steady release of the orange oil vapour to inhibit fungal growth and improve mangosteen quality during storage.

EXPERIMENTAL

Rubberwood (*Hevea brasiliensis*) Sawdust

Rubberwood sawdust obtained from Nakorn Sri Parawood Co., Ltd. (Nakhon-Si-Thammarat province, Thailand) was dried in an air oven at 70 °C for 3 h (moisture content ~3%). Then, it was separated by passing it through a number 20 stainless steel mesh sieve (Endecotts Ltd., London, UK) and stored in a desiccator before use.

Orange Oil and Emulsion Preparation

The orange peel oil derived through steam distillation was provided by Thai-China Flavors and Fragrances Industry Co., Ltd., Bangkok, Thailand. A 500 $\mu\text{L mL}^{-1}$ emulsion was prepared by mixing orange oil with distilled water using a Tween[®] 80 solution

concentration of 3% v/v (Tariko Co., Ltd., Bangkok, Thailand) as the surfactant. The emulsion was homogenised using a T25 Digital Ultra-Turrax® (IKA, Staufen, Germany) at $24,000 \times g$ for 20 min.

Essential oil Absorption Capacity and Sachet Preparation

The rubberwood sawdust (1 g) was dipped in the orange oil emulsions for 300 min to investigate the performance of the rubberwood sawdust and calculate the orange oil absorption capacity. The weight of the sawdust was measured before (W_0) and immediately after (W_t) its removal from the emulsions. The emulsion absorption capacity was determined using Eq. 1,

$$C \text{ (g/g)} = (W_t - W_0) / W_0 \quad (1)$$

where W_t is the weight (g) of rubberwood sawdust at a given immersion time and W_0 is the initial weight of the rubberwood sawdust.

For the sachet preparation, the rubberwood sawdust containing orange oil emulsion was prepared by soaking the emulsion in rubberwood sawdust absorbent at the specific ratio, which provided a maximum emulsion absorption capacity. After drying for 30 min in cool air, the absorbent rubberwood sawdust containing orange oil emulsion in the amounts 70 g, 140 g, 210 g, and 280 g was packed into individual wood pulp paper sachets (10 cm wide \times 15.5 cm long \times 1 cm high; 26 gsm) before testing.

Mangosteen (*Garcinia mangostana*)

Mangosteens with a pink blush on a yellow background (ripening stage 2) were harvested at a temperature of 30 ± 2 °C and relative humidity (RH) of approximately 80% from a local farm in Lan Saka district, Nakhon Si Thammarat province, Thailand. Healthy fruits with uniform colour and size were selected. After arriving at the laboratory, the mangosteens were quickly soaked in distilled water for 5 min at room temperature for cleaning the surface, and the surface was dried for 30 min at room temperature. They were used immediately for the experiments.

Transportation and Storage Conditions

The mangosteens weighed at 15 kg per treatment (repeated three times with three replicates each time) were packed into a corrugated paper box (approximately 35 L, 30 cm wide \times 35 cm long \times 33 cm high) (Nawarat Paper Box Co., Ltd., Bangkok, Thailand). Each of the sachets with 70 g, 140 g, 210 g, and 280 g of the rubberwood sawdust absorbent containing orange oil emulsion was placed in a paper box. This part of the study was carried out based on the transportation condition of mangosteens from the farm in Lan Saka District, Nakhon Si Thammarat, Southern Thailand (Latitude: $8^\circ 24' 22.72''$ N; Longitude: $99^\circ 46' 4.33''$ E) to the fruit market (Talaad Thai) in Pathum Thani, Central Thailand (Latitude: $14^\circ 4' 50.65''$ N; Longitude: $100^\circ 37' 22.95''$ E). Subsequently, the box was closed and transported by truck (Kerry Express (Thailand) Pub Co., Ltd.) from the Nakhon Si Thammarat province to the Pathum Thani province, a distance of 1,598 km ($799 \text{ km} \times 2$) at ambient temperature (approximately 30 ± 2 °C) and 80% RH. The transportation time was three days. After transportation, the fruit was stored continuously for nine days (a total of 12 days).

Effect of Orange Oil Vapour on the Ripening of Mangosteens in a Paper Box *Evaluation of ripening*

The mangosteens in the box were arranged in five treatments as follows: (i) control, (ii) with the sachet of orange oil absorbent at 70 g (iii) with the sachet of orange oil absorbent at 140 g (iv) with the sachet of orange oil absorbent at 210 g, and (v) with the sachet of orange oil absorbent at 280 g. The ripening stage of the fruit was determined using a scale of values between 0 and 6 (stage 0 being yellowish-white, stage 1 being greenish-yellow, stage 2 being pink blush on yellow background, stage 3 being pink, stage 4 being red, stage 5 being reddish-purple, and stage 6 being dark purple) (Jarimopas *et al.* 2009). The ripening scale was recorded on days 0, 3, 6, 9, and 12 after transportation and during storage.

Due to the results from this section, the mangosteens in the box with 210 g orange oil and the one with the control group were selected for colour change, pigments, total phenolic content (TPC), and antioxidant properties.

Colour changes of mangosteens

The colour of the fruit rind was measured at three evenly distributed equatorial sites of each fruit using a colorimeter (ColorFlex, Hunter Associates Laboratory, Virginia, USA). CIELAB colour coordinates were used to determine the degree of lightness (L^* , 0 to 100, black to white), redness-greenness (+ or $-a^*$), and yellowness-blueness (+ or $-b^*$). ΔE values, which represent a measure of the total colour difference, were calculated using Eq. 2,

$$\Delta E = [(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2]^{1/2} \quad (2)$$

where ΔL^* , Δa^* , and Δb^* represents the difference in the L^* , a^* , b^* values at a particular interval from the respective initial values.

Effect of orange oil vapour on pigments of mangosteens

The pigments in the fruit (chlorophyll, carotenoid, and anthocyanin) were examined on day 6. For this, 0.5 g of fruit peel powder was extracted with aqueous acetone (80% v/v) three times. The solution was then centrifuged (ScanSpeed 1580 MGR, LaboGene Co. Ltd., Bjarkesvei, Denmark) at 10,000 g for 20 min. Chlorophyll *a*, *b*, and carotenoid absorbance were then recorded at $\lambda = 663$ nm, $\lambda = 645$ nm, and $\lambda = 470$ nm, respectively, using a spectrophotometer (1205 Vis Spectrophotometer Unico Instrument Co., Ltd., China). The chlorophyll and total carotenoid content were calculated according to a method reported by Thakur *et al.* (2019). The anthocyanin content was determined using the pH differential method. Anthocyanins in the peel powder were extracted three times in the dark with 5 mL of extraction solution (0.05% HCl in methanol). The total supernatant was diluted with the extraction solution to 15 mL and mixed. Subsequently, 1 mL of supernatant was mixed with 4 mL buffers, with pH at 1.0 and 4.5 respectively, and then the mixed solutions were measured at 510 and 700 nm using a spectrophotometer. The anthocyanin content was calculated according to the method described in Lee *et al.* (2005).

Total phenolic content (TPC) and antioxidant properties

The peel of mangosteen (5 g) was extracted using 50 mL solution of methanol (80%), before being placed in a homogeniser tube. The mixture was homogenised and centrifuged for 10 min at 10,000 g. A Whatman no. 1 filter paper was used to filter the supernatant. The TPC of the peel was determined using the Folin–Ciocalteu method. Gallic

acid was used to plot the standard curve. The results were expressed as $\mu\text{g GAE L}^{-1}$ on a fresh weight basis. The 1,1-diphenyl-2-picrylhydrazyl radical scavenging ability (DPPH) of mangosteen peel extracts was evaluated using spectrophotometry. The 2,2'-Azino-Bis-3-ethylbenzothiazoline-6-sulphonic acid (ABTS) free radical scavenging ability was evaluated. The ABTS and DPPH units of the samples were expressed as ascorbic equivalent antioxidant capacity. The results were given as mmol kg^{-1} in fresh weight. All the experiments were performed in triplicate (Cömert *et al.* 2020).

The Effect of Orange Oil in Paper Box on the Mould Surface of Mangosteen

The appearance of mould on the surface of mangosteen

The mangosteens in the box were arranged in five treatments as follows: (i) control, (ii) with the sachet of orange oil absorbent at 70 g, (iii) with the sachet of orange oil absorbent at 140 g, (iv) with the sachet of orange oil absorbent at 210 g, and (v) with the sachet of orange oil absorbent at 280 g. During the storage period, the appearance of mould on the surface of the fruit was investigated using a mould appearance scale. The mould was scored based on a scale of values between 0 and 5 according to the severity of mould appearance (0 = no mould growth; 1 = slight; 2 = moderate; 3 = moderately severe; 4 = severe; 5 = extremely severe) (Owolabi *et al.* 2021b). The mould appearance scale was recorded on days 0, 3, 6, 9, and 12 after transportation and during storage.

Microstructure morphology of mangosteens by SEM analysis

The surface morphology of the treated and untreated fruit at day 12 of the storage was observed using a scanning electron microscope (SEM, JEOL JSM-5800 LV, Kyoto, Japan). The specimens were mounted on stubs and coated with gold before observation.

Effect of Orange Oil Vapour on Quality of Mangosteens after Transportation and Storage

The mangosteens in two boxes were selected for this study (due to the result from the previous section): (i) the one with the control group and (ii) the box with 210 g of the sachet of orange oil absorbent.

Determination of weight loss and firmness of fruit

The weight loss of the fruit during transportation and storage was examined. The fruit from each treatment was weighed using a digital balance (model STX2202, Ohaus Corp., New Jersey, USA) on days 3, 6, and 12 of transportation and storage. The weight loss was calculated using Eq. 3.

$$\text{Weight loss (\%)} = ((\text{Initial weight} - \text{Weighing weight}) / \text{Initial weight}) \times 100 \quad (3)$$

The firmness of the fruit was measured using a texture analyser (LR 5K MK4, Lloyd Instrument Co. Ltd, West Sussex, UK) in compression mode. The test was performed using a stainless steel probe of 5 mm diameter to penetrate 25 mm into three random positions of the fruit with a speed of 1 mm s^{-1} , and the mean maximum force was recorded as fruit firmness (N).

Determination of pH, titratable acidity (TA), and total soluble solids (TSS)

A homogeneous fruit juice sample was prepared from mangosteen flesh to measure the pH, TA, and TSS. The pH was measured using a pH meter (model ST3100-F, Ohaus Corp., New Jersey, USA). The TA was measured by titration using a standard sodium hydroxide solution (0.1 N) and using 0.1% phenolphthalein solution as an indicator. The TA value was expressed as a percentage (%) of citric acid using Eq. 4.

$$\text{Citric acid (\%)} = ((\text{mL of NaOH} \times 0.1 \text{ N NaOH} \times \text{milliequivalent factor}) / \text{mL of juice}) \times 100 \quad (4)$$

The TSS content in the fruit juice was measured with a hand refractometer (Master-T, Atago Co. Ltd., Tokyo, Japan).

Component Analysis of Orange Oil and Mangosteens by Gas Chromatography-Mass Spectrometry (GC-MS)

The untreated sample and the sample treated with a 210 g orange oil sachet were selected for analysis. The mangosteen peel and flesh were cut into small pieces and extracted using ethyl acetate (1:1 w/v). Then, they were mixed gently by shaking overnight. Next, they were filtered using filter paper (Whatman No. 1) and centrifuged (ScanSpeed 1580 MGR, LaboGene Co. Ltd., Denmark) at $12,000 \times g$ for 5 min. Subsequently, the ethyl acetate was removed from the supernatant by blowing with nitrogen gas. Analysis was performed using GC/MS (Agilent 5977A, Agilent Technologies Inc., Santa Clara, CA, USA). The Agilent DB-5 column (dimensions 30 m \times 0.25 mm; film thickness 0.25 μm) was used. The injected volume was 1 μL , the split ratio of the column was 500:1, and the injector temperature was set at 270 $^{\circ}\text{C}$. The oven temperature was initially set at 35 $^{\circ}\text{C}$ and then increased to 200 $^{\circ}\text{C}$ at a rate of 10 $^{\circ}\text{C}/\text{min}$, maintained for 5 min, then increased to 260 $^{\circ}\text{C}$ at a rate of 5 $^{\circ}\text{C}/\text{min}$. The components were identified by comparing their mass spectra fragmentation and computer matching using the Wiley 10 and NIST 14 libraries (Database/ChemStation data system).

Statistical Analysis

All the results were expressed as mean \pm standard deviation. One-way ANOVA and Duncan's post hoc test, with $p < 0.05$ being considered statistically significant, were employed in the statistical analysis using Statistica software (StatSoft, Oklahoma, USA).

RESULTS AND DISCUSSION

Emulsion Absorption Capacity of Rubberwood Sawdust

The orange oil emulsion absorption capacity of rubberwood sawdust is shown in Fig. 1. The rubberwood sawdust was able to absorb the orange oil emulsion with a maximum level of 0.5 g/g at the equilibrium point (120 min). The result of this study corresponds with that of Parichanon *et al.* (2021), where they reported that the rubberwood sawdust could also absorb lime oil emulsion and *Litsea cubeba* oil emulsion at 30% to 40%. This is confirmed to produce an orange oil sachet by dipping the sawdust for 120 min in orange oil emulsion. It is an easy way to produce an active essential oil sachet for placing into the fruit paper box during transportation.

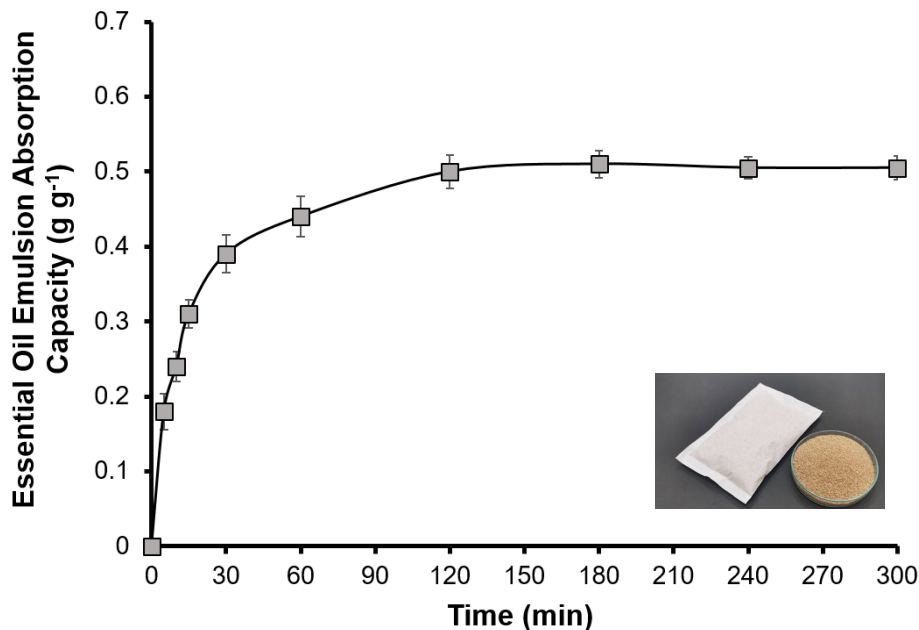


Fig. 1. Orange oil emulsion absorption capacity of rubberwood sawdust (Note: Error bars indicate standard deviation (S.D.))

Effects of Orange Oil Vapour on Ripening During Transportation and Storage

The effects of rubberwood sawdust containing orange oil on the post-harvest ripening of mangosteens are shown in Fig. 2. After transportation and storage for six days, it was found that the sachet containing 210 g of orange oil absorbent delayed the ripening from stage 2 – pink blush on yellow background to stage 4 – red on day 6 and delayed it even further to stage 5 on day 12. This result clearly indicates that the ripening of mangosteens was reduced with an increased amount of orange oil absorbent; however, it was the highest amount that delayed the ripening without a negative effect. Therefore, the optimal concentration of orange oil absorbent in the sachet in the paper box was 210 g. Therefore, 210 g of orange oil absorbent was selected to be compared with the control treatment in the next section of colour and pigment measurement.

The colour of mangosteens with 210 g of orange oil absorbent and control was measured using a chromameter as CIE L^* a^* b^* and ΔE values, as shown in Table 1. The ripening scale was found to be related to the colour change on the peel. Overall, total colour change (ΔE^*) showed a high correlation with the a^* value on the fruit peel, which is associated with the ripening stage and amount of orange oil after transportation and during storage. For control, the stage 2 (pink blush on yellow background) colour of the pericarp of mangosteens changed gradually to stages 3 to 4 (pink-red) within three days after transportation; a fast ripening process with an increase in the value of a^* within six days after transportation and storage. This corresponded to the ripening scale with a value of 6, with the colour changing to a dark purple from day 6 to day 12. For the treatment group, the result demonstrated that regarding the delay in change in the pericarp colour after treatment, from stage 2 to stages 3–4 (pink-red), the a^* value found was lower than that of the control at six days of storage, and delay in reaching stage 5 (reddish) at day 12 correlates well with the decrease in the values of L^* and b^* .

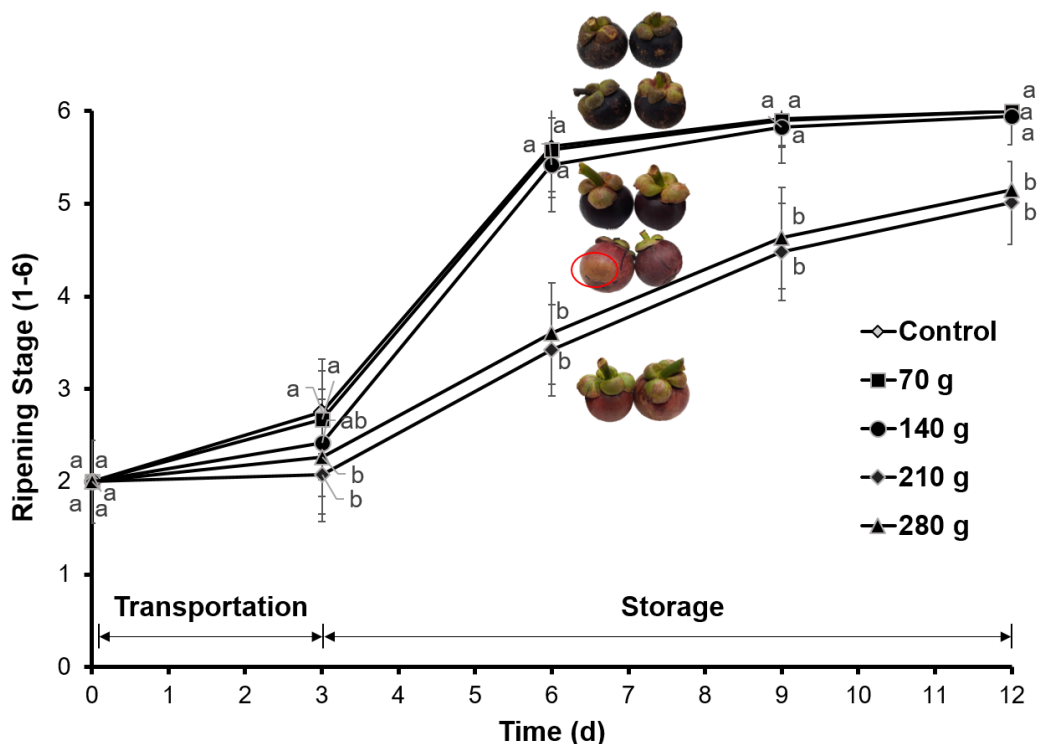


Fig. 2. Ripening of mangosteens and appearance of mangosteens following treatment with sachets containing 70 g, 140 g, 210 g, and 280 g of orange oil absorbent or control (without orange oil treatment). (Note: Error bars indicate standard deviation (S.D.), and “a” and “b” different lower-case letters are significantly different ($p < 0.05$))

The ripening stage of mangosteens is related to the a^* value (redness-greenness), and increased ripening results in increased redness (a^* value). The redness of the pericarp of mangosteens is due to the development of anthocyanin pigments (Parijadi *et al.* 2018). Color of fruits may be attributed to anthocyanin degradation and formation of browning pigments. The colour of mangosteens post-harvest can develop into an intense purple, which is controlled by the regulation of enzymes and genes associated with the anthocyanin biosynthesis pathway (Palapol *et al.* 2009). This experiment confirmed that the anthocyanin content in mangosteens treated with 210 g orange oil did not increase during storage for six days ($1.32 \pm 0.07 \text{ g kg}^{-1}$), compared to the control ($1.96 \pm 0.09 \text{ g kg}^{-1}$). Moreover, chlorophyll ($0.055 \pm 0.003 \text{ g kg}^{-1}$) and carotenoid ($45.98 \pm 0.38 \text{ g kg}^{-1}$) in the treated fruit were significantly higher than in the control chlorophyll ($0.048 \pm 0.003 \text{ g kg}^{-1}$ and $43.15 \pm 0.90 \text{ g kg}^{-1}$, respectively). Additionally, shortly after being treated with orange oil, the TPC and antioxidant capacity (ABTS and DPPH) were found to be significantly increased in the treatment group compared to the control; however, during storage, a decline of TPC, ABTS, and DPPH of the treated fruit was found compared with the control from days 3 to 12 (Table 1). The metabolite accumulation on the fruit peel was significantly different and related to the pigment change. Anthocyanin mainly increased in fruit peel during ripening and accumulated significantly at the mature stage, and the decline of anthocyanin in the fruit treated with 210 g orange oil absorbent was shown to be related to the slow decrease of TPC, ABTS, and DPPH on day 6 and 12. Normally, ethylene signaling networks increase anthocyanin as the fruit changes color to red, purple, and blue. Therefore, it was expected that anthocyanin levels would also vary with the ripening of the

fruit, whose cell-wall breakdown causes the decrease in chlorophyll and carotenoids when the fruit is ripe. However, in this study, the TPC and antioxidant capacity regulation related to the ripening and delayed metabolic color changes of the fruit were found to be varied.

Table 1. Effects of Orange Oil Vapour on the Colour, Total Phenolic Content (TPC), ABTS, and DPPH of the Mangosteens During Storage for 12 d

| Parameters | Transportation and Storage Time (d) | | | | | |
|-------------------------------|-------------------------------------|-----------------------------|-----------------------------|-----------------------------|-----------------------------|-----------------------------|
| | 3 | | 6 | | 12 | |
| | Control | Orange Oil | Control | Orange Oil | Control | Orange Oil |
| Colour | | | | | | |
| -L* | 55.20 ± 5.66 ^{ya} | 58.93 ± 4.92 ^{xa} | 35.12 ± 8.12 ^{yb} | 48.39 ± 5.15 ^{xb} | 19.72 ± 4.98 ^{yc} | 35.18 ± 5.02 ^{xc} |
| -a* | 4.94 ± 1.46 ^{xc} | 3.52 ± 1.70 ^{yc} | 16.03 ± 2.33 ^{xb} | 6.33 ± 1.81 ^{yb} | 27.51 ± 4.01 ^{xa} | 18.10 ± 3.56 ^{ya} |
| -b* | 30.01 ± 3.93 ^{ya} | 33.45 ± 3.55 ^{xa} | 14.64 ± 4.74 ^{yb} | 24.35 ± 4.20 ^{xb} | 7.35 ± 2.78 ^{yc} | 15.43 ± 2.77 ^{xc} |
| ΔE* | 9.67 ± 3.21 ^{xc} | 6.22 ± 3.68 ^{yc} | 36.72 ± 7.79 ^{xb} | 18.64 ± 6.11 ^{yb} | 56.45 ± 3.40 ^{xa} | 37.31 ± 3.69 ^{ya} |
| TPC (g GAE L ⁻¹) | 0.033 ± 0.004 ^{ya} | 0.045 ± 0.002 ^{xa} | 0.016 ± 0.003 ^{yb} | 0.022 ± 0.002 ^{xb} | 0.012 ± 0.002 ^{yc} | 0.017 ± 0.003 ^{xc} |
| ABTS (mmol kg ⁻¹) | 21.09 ± 0.03 ^{ya} | 22.70 ± 0.02 ^{xa} | 13.01 ± 0.02 ^{yb} | 20.37 ± 0.01 ^{xb} | 12.90 ± 0.03 ^{yc} | 13.02 ± 0.01 ^{xc} |
| DPPH (mmol kg ⁻¹) | 8.57 ± 0.01 ^{ya} | 9.12 ± 0.01 ^{xa} | 4.43 ± 0.01 ^{yb} | 6.67 ± 0.01 ^{xb} | 4.32 ± 0.01 ^{yc} | 5.51 ± 0.01 ^{xc} |

Note: Values are presented as average ± standard deviation (S.D.); ^{x-y} different superscripts are significantly different between treatments ($p < 0.05$), and ^{a-c} different superscripts are significantly different between the day of storage ($p < 0.05$).

Orange peel is an important source of orange oil, which, in turn, is an important source of phenolic compounds such as flavonoids, hesperidin, narirutin, tangeritin, naringenin, hesperetin, and diosmetin, which contain antioxidants having beneficial effects on human health (Angoy *et al.* 2020). The essential oil vapour could be used to treat fresh mangosteens to transport them over long distances without affecting their colour quality if treated carefully with the right amount of orange oil in the paper boxes. The current results confirm that using orange oil during storage delayed the ripening of mangosteens, with the advantage of a slightly longer shelf life than the fruit harvested in later stages. This suggests that the application of orange oil vapour that delays the ripening of mangosteens is beneficial for harvesting fruit in stages 1 to 2 for export. The results for ripening showed that the sachet containing 210 g of orange oil absorbent should be selected when studying the shelf life of mangosteens.

Effect of Orange Oil Vapour on Mould Spoilage of Mangosteens

This study investigated the antifungal properties of rubberwood sawdust containing orange oil and its application in a paper packaging box of mangosteens (Fig. 3). The results clearly demonstrated that the sachets containing orange oil absorbent in the paper box at an amount of 210 to 280 g inhibited the growth of natural mould on the surface of mangosteens until 12 days (scale 0 meaning no mould growth). Moreover, the paper boxes containing the sachets with between 70 and 140 g orange oil absorbent provided some antifungal capacity and functional properties compared to the control (without orange oil

treatment). The mould scale on the mangosteens when adding a sachet containing 140 g of orange oil absorbent was less than half that of the control mangosteens, with approximately 1.5 (slight to moderate) on the scale, while the mould scale of mangosteens using a sachet containing 70 g of orange oil absorbent was slightly different from the control with around 2 (moderately) on day 9 and greater than 3 (moderately severe) on day 12.

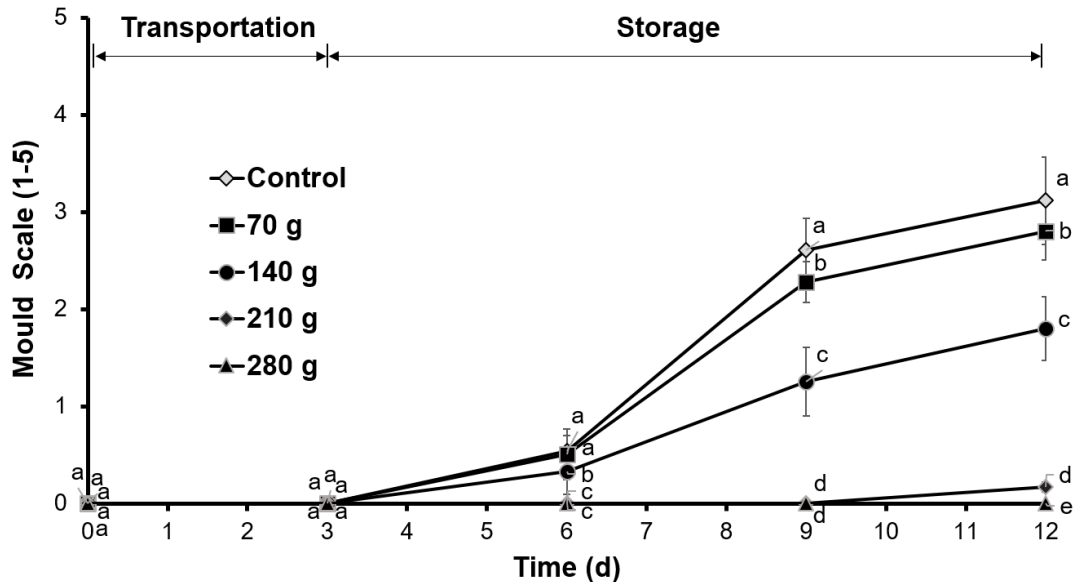


Fig. 3. Mould scale in mangosteens following treatment with sachets containing 70 g, 140 g, 210 g, and 280 g of orange oil absorbent or control (without orange oil treatment). (Note: Error bars indicate standard deviation (S.D.), and ^{a-e} Different superscripts are significantly different ($p < 0.05$))

The SEM results in Fig. 4a-b show that treatment with the 210 g orange oil sachet significantly ($p < 0.05$) impacted the growth of natural mould on the surface of mangosteens. The vapour of orange oil in a paper box significantly reduced the growth of mycelium on the surface of mangosteens.

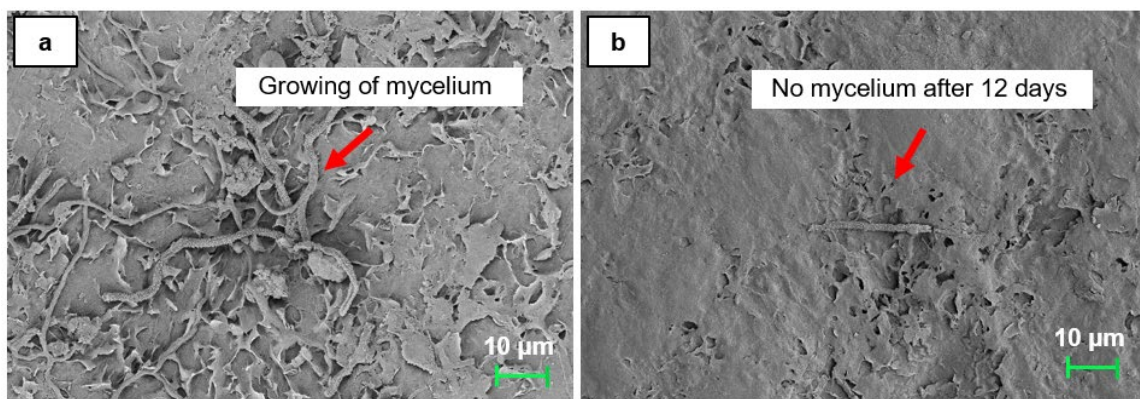


Fig. 4. Scanning electron microscopy of the surface of mangosteens at 1.0 K X; control (a), treated sample (b)

The surface of the control fruit was covered with the mycelium of natural mould which was developed from the hyphae (Fig. 4a), while the surface of the treated

mangosteen was not found with the mycelium growth. The mould was unable to germinate the mould spore on the surface of the mangosteen for the 12 days of testing (Fig. 4b).

This study demonstrated the antifungal potential of orange oil in the vapour phase at 30 ± 2 °C, which could help inhibit mould during transportation at room temperature, and it was demonstrated that in a box of fruit (15 kg) containing orange oil, the surface of the mangosteens had no growth of mould, which usually grows very quickly outside of cold storage. Therefore, this study presents an alternative technique for fruit control inside food packaging that is suitable for transporting and storing fruit without refrigeration. The current result is similar to that of Phothisuwan *et al.* (2021), who found that orange oil could inhibit mould growth on salacca in a closed system and could extend the shelf life of salacca from 10 days (control open air) to at least 28 days. Orange oil contains rich bioactive compounds which function against moulds, but few are applied in food packaging, including this research. The problem of the orange oil degrading quickly and controlling the release of compounds in orange oil may be hard to address in food packaging. Encapsulated orange oil technique for controlling the release of orange oil was used to protect potato slices from natural fungi spoilage (Shi *et al.* 2018). Additionally, more technical applications of orange oil were applied in films (Gucbilmez *et al.* 2019).

Effect of Orange Oil Vapour on Quality of Mangosteens

The quality of mangosteens after transportation and storage is shown in Table 2. The ripening of mangosteen was related to increased weight loss and pH but decreased firmness and TA. After transportation and storage for six days, the weight losses of the treated fruit (2.1%) were significantly lower than those of the control (2.7%). Additionally, after 12 days of storage at 30 ± 2 °C, more decreases in weight loss with reductions of 3.1% in the control and 2.5% in the treated fruit were found, leading to lower visual dehydration of the treated fruit. The loss of weight is well known critical index for shelf-life evaluation of fruit during the post-harvest stage, namely transportation. In this experiment, the orange oil vapour on the surface of the fruit significantly reduced the weight loss in the fruit when helping to delay the ripening of the fruit. However, a decrease in the firmness and TA ($p < 0.05$) in both the control and treated fruit was found. Firmness was measured three days after transportation; for the control, it was 3.7 N and then declined dramatically to nearly 2.8 N and 2.2 N over the climacteric stage for 6 and 12 days, respectively, marking a reduction of nearly 40%. The softening of mangosteen emission was already established during ripening.

On the other hand, the firmness of the treated fruit on day 6 was 2.5 N, lower than that of the control. However, no significant differences were noted between the control and treatment groups after 12 days of storage. Titratable acidity was expressed as the percentage of citric acid equivalents. The levels of TA would be decreased over the ripening stages until stage 6 – dark purple, but the pH, the major source of sourness in the fruit, was higher. The TSS level of mangosteen slightly increased again at stage 4 until stage 6 was reported (Jamil *et al.* 2021), but this result found no change of TSS from day 3 to day 12 between control and treatment groups. The delayed ripening of mangosteen may not be related to the TSS result. Overall, from the results noted after the transportation and storage of mangosteens, a sachet containing 210 g of orange oil absorbent releasing vapour demonstrated great potential for use in maintaining the overall quality for at least 12 days compared with the control.

Table 2. Effects of Orange Oil on the Weight Loss, Firmness, pH, Titratable Acidity, and Total Soluble Solids of the Mangosteens During Storage for 12 d

| Parameters | Transportation and Storage Time (d) | | | | | |
|--------------------------|-------------------------------------|--------------------------|--------------------------|--------------------------|--------------------------|--------------------------|
| | 3 | | 6 | | 12 | |
| | Control | Orange Oil | Control | Orange Oil | Control | Orange Oil |
| Weight loss (%) | 1.9 ± 0.2 ^{xc} | 1.2 ± 0.1 ^{yc} | 2.7 ± 0.2 ^{xb} | 2.1 ± 0.3 ^{yb} | 3.1 ± 0.2 ^{xa} | 2.5 ± 0.2 ^{ya} |
| Firmness (N) | 3.7 ± 0.6 ^{xa} | 3.5 ± 0.5 ^{xa} | 2.8 ± 0.4 ^{xb} | 2.5 ± 0.5 ^{xb} | 2.2 ± 0.4 ^{xc} | 2.2 ± 0.4 ^{xc} |
| pH | 3.0 ± 0.0 ^{xc} | 3.0 ± 0.0 ^{xc} | 3.3 ± 0.1 ^{xb} | 3.2 ± 0.0 ^{yb} | 3.4 ± 0.1 ^{xa} | 3.3 ± 0.1 ^{ya} |
| Titratable acidity (%) | 0.6 ± 0.0 ^{xa} | 0.6 ± 0.1 ^{xa} | 0.5 ± 0.1 ^{yb} | 0.6 ± 0.1 ^{xa} | 0.4 ± 0.0 ^{yc} | 0.5 ± 0.1 ^{xb} |
| Total soluble solids (%) | 14.1 ± 0.5 ^{xa} | 14.0 ± 0.5 ^{xb} | 13.8 ± 0.8 ^{xb} | 14.0 ± 0.7 ^{xb} | 14.2 ± 0.8 ^{xa} | 14.4 ± 0.5 ^{xa} |

Note: Values are presented as average ± standard deviation (S.D.); ^{x-y} different superscripts are significantly different between treatments ($p < 0.05$), and ^{a-c} different superscripts are significantly different between the day of storage ($p < 0.05$)

Possible Mechanism of Action of Orange Oil to Delay the Ripening and Mould Spoilage of Mangosteens

The results obtained confirmed that orange oil is a good inhibitor to delay ripening and against mould on the surface of mangosteens. In the GC-MS results, a total of 13 different compounds constituting 94.12% of orange oil were identified. The GC-MS results showed that d-limonene (88.87%) as monoterpene was the only main major compound of orange oil in this test that was found in both the peel and the flesh of mangosteens (Fig. 5). This result is similar to that of Maróstica and Pastore (2005), who reported that limonene is the major compound of orange oil; however, the percentage of limonene is significantly different due to seasonal variation, geographical conditions, and extraction method. Additionally, d-limonene and myrcene, the major terpenes in citrus, have been monitored in grapefruit in the main emitted by ripe pericarp (Flamini and Cioni 2010). D-limonene could help reduce the respiration of mangosteen (Jamil *et al.* 2021). In addition, it has antioxidant and antimicrobial properties, with which they could interact with free radicals and prevent oxidation (Bacanli *et al.* 2015; Duarte *et al.* 2016). Therefore, the result is the delayed ripening of treated mangosteen.

Regarding antifungal activity, these results are consistent with those of Cai *et al.* (2019), where limonene showed a potent antifungal effect against *Zygosaccharomyces rouxii*. This can be attributed to the disruption of the cell membrane integrity and permeability, which may cause irreversible damage to the cell wall and membrane. Further, Cai *et al.* (2019) showed that limonene exerted a strong inhibitor effect of cellulase activity, which reduced the degree of methylesterification of pectins and influenced the fungi development of the cell wall as well as membrane integrity and permeability. Limonene was evaluated against four plant pathogenic fungi, *Rhizoctonia solani*, *Fusarium oxysporum*, *Penicillium digitatum*, and *Aspergillus niger*, using the mycelial growth-inhibitory technique (Marei *et al.* 2012). Therefore, limonene could be a major inhibitor against mould in this experiment. The results showed that ripening in mangosteens is triggered by the amount of orange oil applied post-harvest and after transportation and storage to the market, with no effect on the ultimate fruit quality. This gives growers a window for handling and retailing the fruit. The fruit also provides a model for further investigation into the control of red colour in relation to antioxidant capacity and other ripening stimulants.

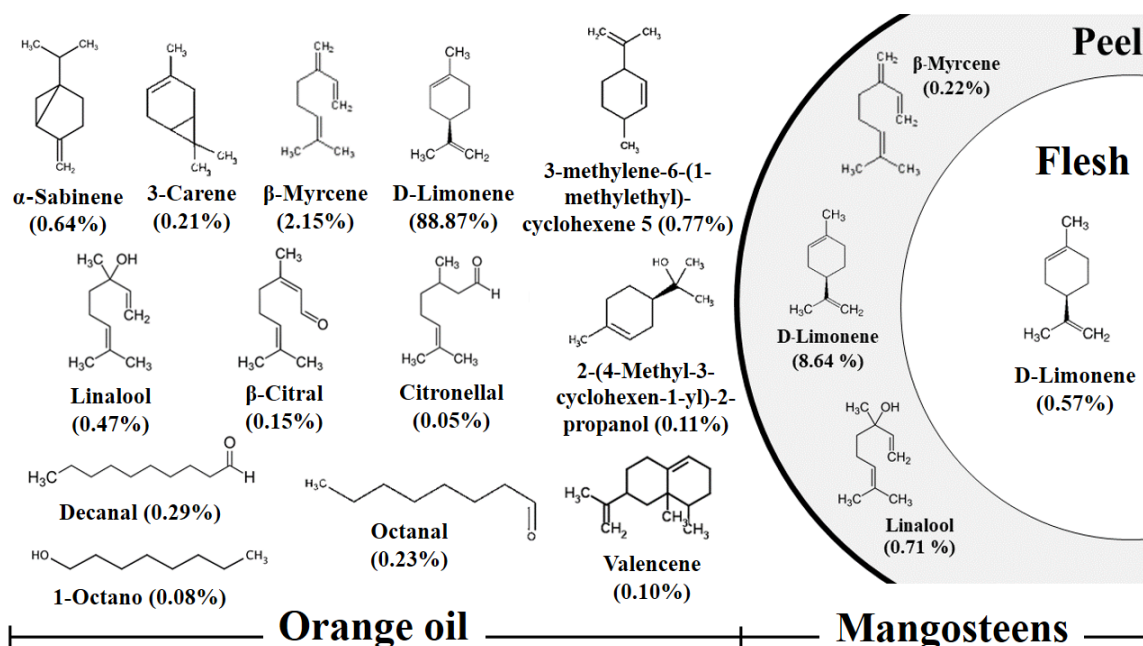


Fig. 5. The components present in orange oil and the peel and flesh of treated and untreated mangosteens (control) at day 12 of the storage period

CONCLUSIONS

1. The application of a sachet of rubberwood sawdust containing orange oil at a concentration of 210 g in corrugated paper box packaging (35 L) for the transportation and storage of mangosteens (12 days at 30 ± 2 °C, 80% RH) had significant effects on the quality and appearance of the fruit.
2. The treated mangosteens revealed a reduction in mould decay, ripening, changes in colour, and weight loss, and the fruit had a longer shelf life compared with the control.
3. Limonene in orange oil may have played a key role in inhibiting the mycelial growth of natural mould on the surface of the treated mangosteens.
4. Orange oil helped to reduce the development of anthocyanin pigments and reduce the degradation of chlorophyll pigments in mangosteens during storage, which delayed the ripening and maintained the quality of the mangosteens.

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REFERENCES CITED

- Aizat, W. M., Ahmad-Hashim, F. H., and Jaafar, S. N. S. (2019). "Valorization of mangosteen, 'The Queen of Fruits,' and new advances in postharvest and in food and engineering applications: A review," *J. Adv. Res.* 20, 61-70. DOI: 10.1016/j.jare.2019.05.005
- Angoy, A., Ginies, C., Goupy, P., Bornard, I., Ginisty, P., Sommier, A., Valatd, M., and Chemat, F. (2020). "Development of a green innovative semi-industrial scale pilot combined microwave heating and centrifugal force to extract essential oils and phenolic compounds from orange peels," *Innov. Food Sci. Emerg.* 61, 102338. DOI: 10.1016/j.ifset.2020.102338
- Bacanlı, M., Başaran, A. A., and Başaran, N. (2015). "The antioxidant and antigenotoxic properties of citrus phenolics limonene and naringin," *Food Chem. Toxicol.* 81, 160-170. DOI: 10.1016/j.fct.2015.04.015
- Caccioni, D. R., Guizzardi, M., Biondi, D. M., Renda, A., and Ruberto, G. (1998). "Relationship between volatile components of citrus fruit essential oils and antimicrobial action on *Penicillium digitatum* and *Penicillium italicum*," *Int. J. Food Microbiol.* 43(1-2), 73-79. DOI: 10.1016/S0168-1605(98)00099-3
- Cai, R., Hu, M., Zhang, Y., Niu, C., Yue, T., Yuan, Y., and Wang, Z. (2019). "Antifungal activity and mechanism of citral, limonene and eugenol against *Zygosaccharomyces rouxii*," *Lwt* 106, 50-56. DOI: 10.1016/j.lwt.2019.02.059
- Cömert, E. D., Mogol, B. A., and Gökmen, V. (2020). "Relationship between color and antioxidant capacity of fruits and vegetables," *Curr. Res. Nutr. Food Sci.* 2, 1-10. DOI: 10.1016/j.crf.2019.11.001
- Duarte, A., Luís, Á., Oleastro, M., and Domingues, F. C. (2016). "Antioxidant properties of coriander essential oil and linalool and their potential to control *Campylobacter* spp.," *Food Control* 61, 115-122. DOI: 10.1016/j.foodcont.2015.09.033
- Flamini, G. and Cioni, P. L. (2010). "Odour gradients and patterns in volatile emission of different plant parts and developing fruits of grapefruit (*Citrus paradisi* L.)," *Food Chem.* 120(4), 984-992. DOI: 10.1016/j.foodchem.2009.11.037
- Gorinstein, S., Poovarodom, S., Leontowicz, H., Leontowicz, M., Namiesnik, J., Vearasilp, S., Haruenkit, R., Ruamsuke, P., Katrich, E., and Tashma, Z. (2011). "Antioxidant properties and bioactive constituents of some rare exotic Thai fruits and comparison with conventional fruits: *in vitro* and *in vivo* studies," *Food Res. Int.* 44(7), 2222-2232. DOI: 10.1016/j.foodres.2010.10.009
- Gucbilmez, C., Oksuz, O., and Arıcı, M. (2019). "Fortification of edible zein films with thyme, laurel and orange essential oils and determination of their antifungal and radical scavenging properties," *J. Food Qual.* 70(3), 82-88. DOI: 10.2376/0003-925X-70-82
- Jamil, I. N., Sanusi, S., Mackeen, M. M., Noor, N. M., and Aizat, W. M. (2021). "SWATH-MS proteomics and postharvest analyses of mangosteen ripening revealed intricate regulation of carbohydrate metabolism and secondary metabolite biosynthesis," *Postharvest Biol. Technol.* 176, 111493. DOI: 10.1016/j.postharvbio.2021.111493
- Jarimopas, B., Pushpariksha, P., and Singh, S. P. (2009). "Postharvest damage of mangosteen and quality grading using mechanical and optical properties as indicators," *Int. J. Food Prop.* 12(2), 414-426. DOI: 10.1080/10942910701837262
- Khewkhom, N., Sangchote, S., and Sungsiri, T. (2013). "Postharvest control of fruit rot

- of mangosteen by plant extracts from *Zingiberaceae* family,” *Acta Hortic.* 973, 119-124. DOI: 10.17660/ActaHortic.2013.973.14
- Lee, J., Durst, R. W., Wrolstad, R. E., Eisele, T., Giusti, M. M., Hach, J., Hofsommer, H., Koswig, S., Krueger, D. A., Kupina, S., Martin, S. K., Martinsen, B. K., Miller, T. C., Paquette, F., Ryabkova, A., Skrede, G., Trenn, U., and Wightman, J. D. (2005). “Determination of total monomeric anthocyanin pigment content of fruit juices, beverages, natural colorants, and wines by the pH differential method: Collaborative study,” *J. AOAC Int.* 88(5), 1269-1278. DOI: 10.1093/jaoac/88.5.1269
- Lerslerwong, L., Rugkong, A., Imsabai, W., and Ketsa, S. (2013). “The harvest period of mangosteen fruit can be extended by chemical control of ripening – A proof of concept study,” *Sci. Hortic.* 157, 13-18. DOI: 10.1016/j.scienta.2013.03.027
- Li, Y., Tang, C., and He, Q. (2021). “Effect of orange (*Citrus sinensis* L.) peel essential oil on characteristics of blend films based on chitosan and fish skin gelatin,” *Food Biosci.* 41, 100927. DOI: 10.1016/j.fbio.2021.100927
- Marei, G. I. K., Rasoul, M. A. A., and Abdelgaleil, S. A. M. (2012). “Comparative antifungal activities and biochemical effects of monoterpenes on plant pathogenic fungi,” *Pestic. Biochem. Phys.* 103(1), 56-61. DOI: 10.1016/j.pestbp.2012.03.004
- Maróstica Jr, M. R. and Pastore, G. M. (2007). “Production of R-(+)- α -terpineol by the biotransformation of limonene from orange essential oil, using cassava waste water as medium,” *Food Chem.* 101(1), 345-350. DOI: 10.1016/j.foodchem.2005.12.056
- Mazlan, M. A. F., Uemura, Y., Yusup, S., Elhassan, F., Uddin, A., Hiwada, A., and Demiya, M. (2016). “Activated carbon from rubber wood sawdust by carbon dioxide activation,” *Procedia Eng.* 148, 530-537. DOI: 10.1016/j.proeng.2016.06.549
- Owolabi, I. O., Songsamoe, S., Khunjan, K., and Matan, N., (2021a). “Effect of tapioca starch coated-rubberwood box incorporated with essential oils on the postharvest ripening and quality control of mangosteen during transportation,” *Food Control* 126, article no. 108007. DOI: 10.1016/j.foodcont.2021.108007
- Owolabi, I. O., Songsamoe, S., and Matan, N. (2021b). “Combined impact of peppermint oil and lime oil on Mangosteen (*Garcinia mangostana*) fruit ripening and mold growth using closed system,” *Postharvest Biol. Technol.* 175, article no.111488. DOI: 10.1016/j.postharvbio.2021.111488
- Palapol, Y., Ketsa, S., Lin-Wang, K., Ferguson, I. B., and Allan, A. C. (2009). “A MYB transcription factor regulates anthocyanin biosynthesis in mangosteen (*Garcinia mangostana* L.) fruit during ripening,” *Planta* 229(6), 1323-1334. DOI: 10.1007/s00425-009-0917-3
- Parichanon, P., Matan, N., Limbo, S., D'Incecco, P., and Matan, N. (2021). “Natural cellulosic material characteristics: A possibility to develop antimicrobial active fiber-based packaging,” *BioResources* 16(3), 5450-5466. DOI: 10.15376/biores.16.3.5450-5466
- Parijadi, A. A. R., Putri, S. P., Ridwani, S., Dwivany, F. M., and Fukusaki, E. (2018). “Metabolic profiling of *Garcinia mangostana* (mangosteen) based on ripening stages,” *J. Biosci. Bioeng.* 125(2), 238-244. DOI: 10.1016/j.jbiosc.2017.08.013
- Pedraza-Chaverri, J., Cárdenas-Rodríguez, N., Orozco-Ibarra, M., and Pérez-Rojas, J. M. (2008). “Medicinal properties of mangosteen (*Garcinia mangostana*),” *Food Chem. Toxicol.* 46(10), 3227-3239. DOI: 10.1016/j.fct.2008.07.024
- Phothisuwan, S. and Matan, N. (2021). “Controlled release of essential oil from a rubberwood box using activated carbon to extend the shelf life of strawberries and its possible mode of action,” *BioResources* 16(1), 1726. DOI:

10.15376/biores.16.1.1726-1739

- Phothisuwan, S., Matan, N., and Matan, N. (2021). "The influence of a closed system combining orange oil and mode of action on quality preservation of salacca fruit," *Food Control* 130, 108265. DOI: 10.1016/j.foodcont.2021.108265
- Radünz, M., Camargo, T. M., dos Santos Hackbart, H. C., Alves, P. I. C., Radünz, A. L., Gandra, E. A., and da Rosa Zavareze, E. (2021). "Chemical composition and *in vitro* antioxidant and antihyperglycemic activities of clove, thyme, oregano, and sweet orange essential oils," *LWT* 138, 110632. DOI: 10.1016/j.lwt.2020.110632
- Saengwong-ngam, R., Matan, N., and Matan, N. (2022). "Effects of a plant-cushioning material containing tangerine oil on bruising and mould growth of banana (kluai hom thong) and its potential reuse," *BioResources* 17(1), 37-51. DOI: 10.15376/biores.17.1.37-51
- Shi, Y., Huang, S., He, Y., Wu, J., and Yang, Y. (2018). "Navel orange peel essential oil to control food spoilage molds in potato slices," *J. Food Prot.* 81(9), 1496-1502. DOI: 10.4315/0362-028X.JFP-18-006
- Thakur, R., Pristijono, P., Bowyer, M., Singh, S. P., Scarlett, C. J., Stathopoulos, C. E., and Vuong, Q. V. (2019). "A starch edible surface coating delays banana fruit ripening," *LWT* 100, 341-347. DOI: 10.1016/j.lwt.2018.10.055

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