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Crystallisation behaviour of sunflower and longan honey with glucose addition by absorbance measurement

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Abstract

The present work aimed to study the crystallisation behaviour of honey with glucose addition by using the absorbance measurement at 660 nm. The Avrami model was used to explain the crystallisation kinetic. The effect of glucose addition on the colour and microstructure of crystallised honey was also determined. Sunflower and longan honey were used in the present work. The biochemical compositions of honey samples were analysed before the addition of glucose powder at four concentrations (1.0, 1.5, 2.0, and 2.5%; w/w) and stored at 10 - 15°C. The absorbance at 660 nm, colour, and microstructure were measured until full crystallisation. The results showed that different honey compositions exhibited different crystallisation behaviours, which could be monitored by the absorbance measurement at 660 nm as well. The addition of glucose powder could stimulate the crystallisation time, thus affecting the crystallisation rate. The Avrami equation was found to be a good model for describing the crystallisation behaviour from the intensity of absorbance values. The highest crystallisation rate was found at 2.0% (w/w) glucose addition at the rate constant (k) of 0.177 ± 0.038 and 0.083 ± 0.039 in sunflower and longan honey, respectively. The Avrami index (n) was relevant to the crystallisation rate, but not clearly related to crystal shape or size. The crystals of all glucose-added honey contained small crystals in bouquets. The yellow colour became lighter during storage period. Suitable glucose content and absorbance monitoring at 660 nm could be used to control the crystallisation of honey for obtaining creamed honey product with good texture.

Introduction

Honey is a naturally sweet and viscous food produced by bees and some related insects. It consists of approximately 181 substances including fructose, glucose, water, maltose, and other constituents (Hagr *et al.,* 2017; Singh and Singh, 2018). Its physicochemical properties and flavour depend on the flower sources, geographical regions, and honeybee types. These factors affect the crystallisation behaviours as well. The most popular and abundant kind of honey in Northern Thailand is longan honey (Chaikham *et al.,* 2016), which has a strong fruity scent and unique taste with dark brown colour. Due to the honey's brown colour and strong aroma, it is almost impossible to further develop it into another food product; hence, almost all longan honey found in Thailand markets is pure liquid longan honey. To further increase its market value, longan honey should and could be developed into new food product. One such product which would likely be popular and interesting in Thailand is creamed honey.

Creamed honey product is produced from honey with the crystallisation process controlled, and it is normally produced from honey that is ready to crystallise during storage. However, a coarse and gritty texture is often found due to the crystallised honey being subjected to an uncontrolled crystallisation process. By controlling the crystallisation process and proper raw material preparations, this can result in a value-added honey product. In Thailand, sunflower honey tends to crystallise during storage and might easily be produced into a creamed honey product. Therefore, we selected two honey types to be assessed in the present work, which is sunflower and longan honey, which consisted of different properties.

Dyce method is a method of producing creamed honey, and the process is as follows: 5 - 10% (w/w) of granulated honey is added and stored at 14°C until full crystallisation (Elhamid and Abou-Shaara, 2016). However, many researchers have found different textures when using different honey types. Therefore, the crystallisation behaviour and glucose effect are important parameters for the design of suitable creamed honey processing. Some reports (Conforti

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et al., 2006; Costa *et al.,* 2015; Jaturonglumlert *et al.,* 2015; Elhamid and Abou-Shaara, 2016) have studied the influence of storage temperature and glucose content on crystallisation behaviour and colour of honey (Kędzierska-Matysek *et al.,* 2016a; 2016b). However, no studies have correlated these with creamed honey texture. Moreover, there are few reports that have endeavoured to develop a quick spectrophotometric method to monitor the crystallisation process of creamed honey production from sunflower and longan honey. Several studies (Lupano, 1997; Venir *et al.,* 2010; Laos *et al.,* 2011; Dettori *et al.,* 2018) have been published about the crystallisation kinetics of honey and sugar mixtures using differential scanning calorimetry (DSC), and tried to explain the kinetics of honey crystallisation with the mathematical model.

The Avrami equation was found to suitably describe the honey crystallisation kinetics from DSC measurement during induced crystallisation (Dettori *et al.,* 2018), but has not applied the absorbance value at 660 nm. Conforti *et al.* (2006) and Lupano (1997) reported that turbidity measurement at 660 nm can be taken as an indicator of honey granulation. The increase in turbidity indicates the presence of more honey crystals. But there is a lack of study concerning honey crystallisation modelling, both for monitoring the crystallisation kinetic, and further application in the design of creamed honey process.

Therefore, the objective of the present work was to describe the crystallisation behaviour of honey with glucose addition by measuring the absorbance at 660 nm and Avrami model. The information obtained was applied to design a creamed honey process with the best texture. The effects of glucose addition on the colour and microstructure of crystallised honey were also determined.

Materials and methods

Raw materials and sample preparation

Sunflower and longan honey (fresh and non-crystallised honey) were purchased from Supha Bee Farm in Chiang Mai, Thailand. Samples were heated at 50°C to eliminate natural crystals and osmotolerant/osmophilic yeasts by using a water bath (Tosi *et al.,* 2004). Heated honey samples were kept at room temperature before seeding addition. Glucose monohydrous powder ($C_6H_{12}O_6$, Merck, Germany) was employed as seed (Elhamid and Abou-Shaara, 2016; Dettori *et al.,* 2018). Four levels of glucose were added at 1.0, 1.5, 2.0, and 2.5% (w/w) of ~40 g of liquid honey. Glucose powder was added in both honey types, but none was added to the control sample. Samples were manually stirred with a spatula at room temperature for about 10 min to thoroughly mix between the glucose powder and liquid honey (Dettori *et al.,* 2018). Samples were then stored at a chilling temperature (10 - 15°C) to allow for crystallisation process. The temperature in the refrigerator was daily checked during the experiment. Creamed honey samples were periodically measured for absorbance at different sampling periods and visual observation was conducted to check the overall appearance. If there were crystals occurring throughout the honey sample, it was deemed as full crystallisation.

Biochemical compositions of honey

The moisture content was determined following a refractive index method (Bogdanov, 2009) by measuring the refractive index of honey with a digital Abbe refractometer (KRUSS, Germany). Three samples from each trial were analysed for determination of moisture, and the average moisture content was reported. Sugar content (fructose, glucose, sucrose, and maltose) was determined by HPLC, following the method 977.20 (AOAC, 2000). Water activity was measured by using a water activity meter (AQUA Lab 3TE, USA). The measurements of each sample were duplicated, and the average water activity was calculated.

The colour was measured by using a spectrophotometer (HunterLab Miniscan XE plus, Germany) and reported in values of L*, a*, and b*. Calibration of instrument was done with a black and white standard tile before each set of measurements. The L* colour value presented the degree of brightness or whiteness of the sample, while a* and b* colour values indicated the degree of redness and yellowness, respectively. Three measurements were done in each sample. All parameters were used to calculate the hue angle (h°) and Whiteness Index (WI) (Briones and Aguilera, 2005; Costa *et al.,* 2015) as in Eqs. 1, 2, and 3:

$$h° = \arctan gent\left(b^*/a^*\right)$$

when a* > 0 and b* > 0 (Eq. 1)

$$h° = \arctan gent\left(b^*/a^*\right)+180$$

when a* < 0 (Eq. 2)

$$WI = 100-\left[\left(100-L^*\right)^2 + a^{*2} + b^{*2}\right]^{0.5}$$

(Eq. 3)

Microstructure analysis

Honey samples were microstructurally

observed at room temperature using a light microscopy (Olympus, model BX51TF, Japan) at a magnification of 40× (Lupano, 1997).

Turbidity of honey

Honey samples were poured into a cuvette of about 1 cm path-length. Some trials with many types of liquid honey were taken to scan for wavelength in a range between 200 - 700 nm which presented a smooth curve in a range between 600 - 700 nm. Therefore, the absorbance was measured at 660 nm at room temperature by a spectrophotometer (Model SPECTRO SC, USA) following trials and literature review by Lupano (1997), Conforti *et al.* (2006) and Costa *et al.* (2015).

Crystallisation kinetic of honey

Regarding different samples that reached different final absorbance values (A_{660}), we considered X as 1, which is the maximum absorbance (A_{660max}) to compare kinetics of the crystallisation process. Thus, we considered the increase of the relative crystallised fraction $X(t)$ during storage as Eq. 4.

$$X(t) = \frac{A_{660}}{A_{660_{max}}} \quad \text{(Eq. 3)}$$

where, (A_{660}) = the absorbance wavelength at 660 nm at any time, (A_{660max}) = the absorbance wavelength at 660 nm at a maximum value of any treatment, and $X(t)$ = the relative crystallised glucose fraction at the time t. The Avrami equation (Avrami, 1939) describes the proportion of crystals presented at time (t) compared with glucose crystals in full crystallisation time, as shown in Eq. 5 (Dettori *et al.,* 2018).

$$X(t) = 1 - EXP\left[-k\left(t^n\right)\right] \quad \in [0,1] \quad \text{(Eq. 4)}$$

where, k = rate constant of crystallisation process, and n = Avrami index, a parameter characteristic of nucleation and growth mechanisms of the crystals.

Statistical analysis

As an independent experiment, the results were expressed as mean ± standard deviation (mean ± S.D.) of triplicate testing (n = 3). Statistical analysis was performed using analysis of variance (ANOVA), followed by Duncan's multiple range test (DMRT). Duncan's method was employed to analyse the significant difference in responses at $p \leq 0.05$. Statistical processing was carried out using SPSS 17. The coefficient of determination (R^2) was also calculated to statistically evaluate the accuracy of the mathematical model to simulate crystallisation kinetics.

Results and discussion

Characterisation and biochemical compositions of honey

Both sunflower and longan honey were clear liquid; in which crystals were not present before the addition of glucose. The biochemical compositions of sunflower and longan honey are shown in Table 1 with a corresponding glucose concentration of 39.01 and 33.31%, respectively. The water percentage of sunflower and longan honey were 18.37 and 15.70%, respectively. Thus, the resulting glucose/water (G/W) ratio ranged between 2.11 - 2.12. The fructose/glucose

Table 1. Biochemical compositions (g/100 g) of honey samples.

Honey property	Honey type	
	Sunflower honey	Longan honey
Moisture (% wet basis)	18.37 ± 0.09	15.70 ± 0.31
Total solids (% w/w)	80.06 ± 0.18	82.80 ± 0.17
Water activity (a_w)	0.616 ± 0.002	0.570 ± 0.004
Fructose	40.15 ± 0.57	41.56 ± 2.14
Glucose	39.01 ± 0.51	33.31 ± 1.03
Sucrose	0.39 ± 0.05	0.76 ± 0.03
Maltose	1.50 ± 0.04	0.57 ± 0.04
Lactose	*not detected*	*not detected*
(Fructose/Glucose) ratio	1.03	1.25
(Glucose/Water) ratio	2.12	2.11

Data are means ± standard deviations of three replicates (n = 3).

(F/G) ratio of sunflower honey was less than 1.14 (White, 1975), where rapid crystallisation was found; while F/G ratio of longan honey was 1.25, relatively close to optimised value of 1.33 and categorised as slow or non-granulating honey. Regarding the tendency of honey, crystallisation depends on its composition and moisture content. This is why the sunflower and longan honey were selected in the present work; they represent honey with rapid and non-crystallisation behaviours, respectively. The absorbance measurement during storage was studied in order to explain the crystallisation behaviour. Glucose powder was added to induce crystallisation (Lupano, 1997).

Turbidity of honey

More granulated honey resulted in greater turbidity values. An increase in the absorbance intensity at 660 nm was considered valid to determine the extension of honey granulation (Lupano, 1997). Liquid of sunflower and longan honey, which were stored at chilling temperature, were found to have an increasing turbidity interpreted from the absorbance at 660 nm. From different crystallisation behaviours (Figure 1A), it was shown that the absorbance at 660 nm of honey stored at 10 - 15°C without glucose addition (control sunflower and control longan) is a function of storage time. However, this has shown different characteristics, and the absorbance rapidly increased in sunflower honey until the absorbance value reached 3 (23 days). Moreover, the appearance was in light yellow colour and turbid. The absorbance of longan honey slowly increased and no crystals were found for more than 120 days due to it being slow or non-granulating honey. Therefore, the absorbance at 660 nm was correlated with the overall appearance of honey during storage in chilled temperature.

A correlation between the fructose/glucose (F/G) ratio and crystallisation behaviour was also obtained. Sunflower honey had a low F/G ratio (1.03) which was equal or less than 1.14, resulted in fast or rapid crystallisation (Tosi *et al.,* 2004). It also presented a rapidly increased absorbance at 660 nm. Meanwhile, the F/G ratio of longan honey was higher (1.25) and no crystals were found for more than six months. Therefore, the absorbance curve presented by the crystallisation behaviour of longan honey was slower than that of sunflower honey. Laos *et al.* (2011) reported on the correlation of F/G ratio and the crystallisation time; where a higher F/G ratio presented a longer crystallisation time in eleven Estonian honey samples. These results are similar to Gleiter *et al.* (2006) which mentioned that the required time for honey to crystallise depends on the F/G ratio.

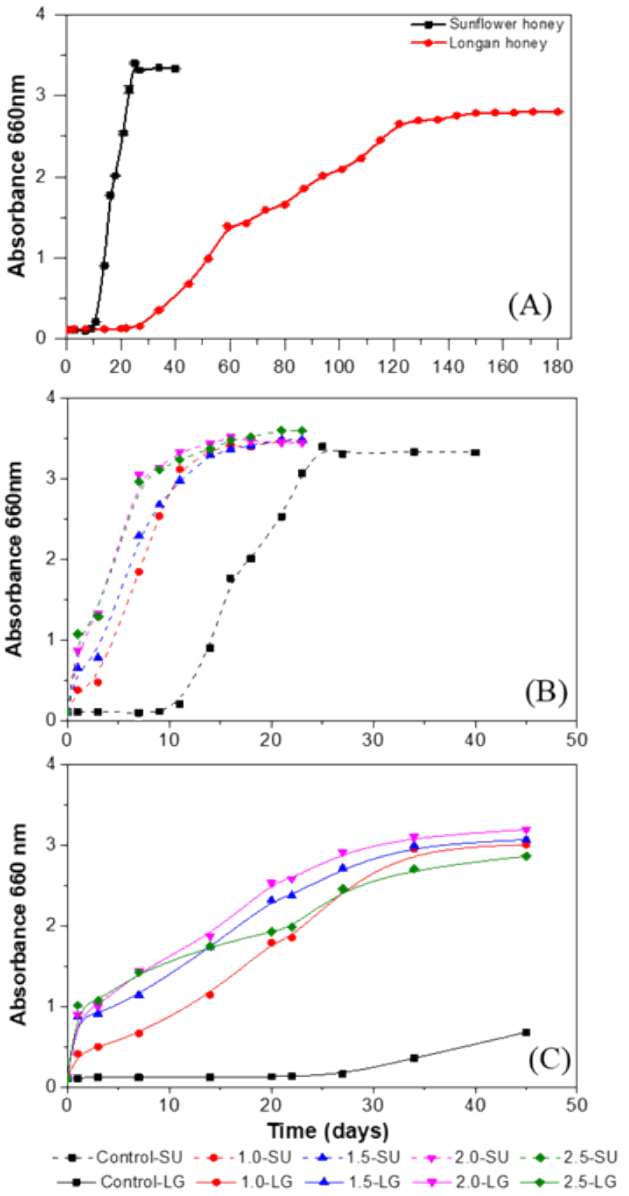

Figure 1. Absorbance at 660 nm of (A) natural crystallisation behaviour, the crystallisation behaviour was induced with glucose addition of sunflower (B), and longan honey (C) as a function of storage time.

Correlation between glucose addition and crystallisation behaviour

A negative correlation was found between glucose addition and the time needed until full crystallisation. An increasing amount of glucose powder added to liquid honey has led to decrease in crystallisation time.

The intensity of absorbance at 660 nm was carried out to monitor the increase in glucose crystal in honey samples during storage at chilling temperature. The results showed that we could accelerate crystallisation by adding glucose powder into both samples (Figure 1B and 1C). When absorbance or turbidity was increased, the crystallisation rate also increased, but with non-linear characteristics. It

rapidly increased in the initial phase, and when close to full crystallisation, the curve continued, particularly in sunflower honey. These results are consistent with a study by Lupano (1997). Crystallisation times of sunflower and longan honey were 39 and 81% faster, respectively, after 1.0% (w/w) of glucose was added. Meanwhile 2.0% (w/w) glucose allowed the highest crystallisation rate, where 70 and 88% were shown to be faster in sunflower and longan honey, respectively (Table 2). Longan honey, which is naturally non-crystallised honey, reached full crystallisation within 22 days after 2.0% (w/w) glucose was added, while sunflower honey only took seven days. These findings differed from the study by Elhamid and Abou-Shaara (2016) who reported that high glucose content leads to high crystallisation. The fastest crystallisation process (27 days) occurred when 1.2, 1.8, or 2.4% (w/w) powder glucose was added into clover and cotton honey. It was faster than 0.3% (66 days) and 0.6% (56 days). The fastest crystallisation time was found in sample with the highest concentration of glucose or 2.4% (w/w). Such different results might be due to the different honey types, geographical areas, and floral sources, which potentially lead to different crystallisation behaviours and the rate of crystallisation.

Modelling the absorbance data with Avrami model

To observe the crystallisation behaviour and compare the evolution of the crystallisation process by using the Avrami model, the equation was simulated to find the value of the crystallisation constant and (k) Avrami index (n). The maximum value of absorbance at 660 nm (A_{660max}) in each treatment was considered as the relative crystallised fraction, or X as 1. We considered an increase of the relative crystallised fraction, $X(t)$, during storage, following Eq. 4. The experimental data and corresponding fitted model of both honey samples are shown in Figure 2. The rate of crystallisation for all treatments in sunflower honey was not linear but seemed to be characterised by an initial fast phase followed by a slower one. The slope of the crystallisation rate plot of sunflower honey was greater after being induced with glucose powder. However, the slope became static when honey samples reached full crystallisation, or the nuclei were in the growth phase. Similar behaviour was observed by Venir *et al.* (2010). The statistical parameters (R^2) of the Avrami model were used to compare between two types of creamed honey. The higher the R^2, the better it fitted with the model and the calculated parameters obtained by modelling the absorbance data with the Avrami model (Table 2). The statistical parameters of the Avrami model are suitable to describe the crystallisation kinetics. R^2 values were high (0.818 - 0.992) in both honey samples, except for the control sample, without glucose addition. This is similar to the Avrami equation used to monitor the crystallisation kinetics of liquid honey with fine crystals added at 14°C to increase the crystallisation rate, which presented R^2 values in the range of 0.924 - 0.931 (Dettori *et al.,* 2018).

The crystallization constant (k) increased with the increase in glucose powder. When we added glucose powder at only 1.0% (w/w), the constant

Table 2. Full crystallisation time, the absorbance value at 660 nm, Avrami parameters (and) with standard deviation (S.D.), and statistical parameters of crystallised honey following the addition of glucose powder at 1.0, 1.5, 2.0, and 2.5 (% w/w) to liquid sunflower and longan honey.

Sample	Treatment	Crystallisation time (day)	Absorbance value at 660 nm		k	n	R^2	Equation
			Day 0	at full crystallisation				
Sunflower honey	Control	23	0.115 ± 0.000[a]	3.072 ± 0.047[a]	$1.082x10^{-5}$ ± 0.000	3.919 ± 0.307	0.991	$X=1-EXP\left(-1.082x10^{-5}t^{3}\right.$
	1.0%	14	0.385 ± 0.001[b]	3.360 ± 0.008[b]	0.016 ± 0.007	2.010 ± 0.196	0.992	$X=1-EXP\left(-0.016t^{2.010}\right)$
	1.5%	14	0.661 ± 0.002[c]	3.303 ± 0.008[c]	0.078 ± 0.022	1.341 ± 0.133	0.986	$X=1-EXP\left(-0.078t^{1.341}\right)$
	2.0%	7	0.877 ± 0.008[d]	3.056 ± 0.005[a]	0.177 ± 0.038	1.159 ± 0.123	0.984	$X=1-EXP\left(-0.177t^{1.159}\right)$
	2.5%	9	1.082 ± 0.002[e]	3.118 ± 0.007[d]	0.212 ± 0.048	1.000 ± 0.117	0.976	$X=1-EXP\left(-0.212t\right)$
Longan honey	Control	180	0.111 ± 0.001[a]	2.454 ± 0.002[a]	-	-	-	-
	1.0%	34	0.412 ± 0.002[b]	2.953 ± 0.004[b]	0.010 ± 0.008	1.529 ± 0.267	0.956	$X=1-EXP\left(-0.01t^{1.529}\right)$
	1.5%	34	0.878 ± 0.003[c]	2.996 ± 0.002[c]	0.074 ± 0.038	1.000 ± 0.182	0.930	$X=1-EXP\left(-0.074t\right)$
	2.0%	22	0.890 ± 0.025[c]	2.580 ± 0.001[d]	0.083 ± 0.039	1.000 ± 0.172	0.937	$X=1-EXP\left(-0.083t\right)$
	2.5%	27	1.010 ± 0.013[d]	2.456 ± 0.003[e]	0.075 ± 0.055	1.000 ± 0.261	0.818	$X=1-EXP\left(-0.075t\right)$

Data are means ± standard deviations of three replicates ($n=3$). Means followed by the different letter are significantly different between mean values ($p \leq 0.05$).

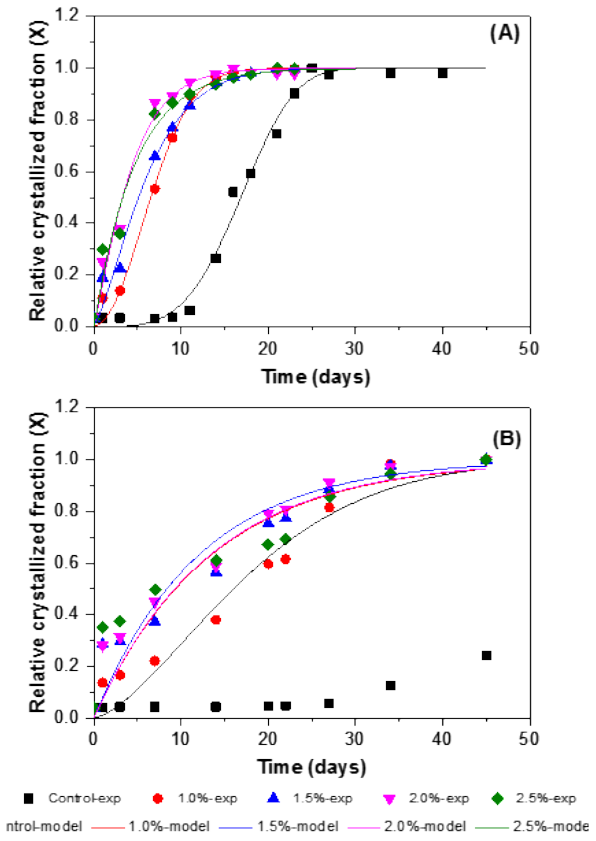

Figure 2. Experiment (exp) and modelled (model) data of relative crystallised fraction *X(t)* obtained with the Avrami equation of (A) sunflower honey, and (B) longan honey with glucose addition (% w/w).

constant *k* (0.016) was higher than the sample without glucose addition (1.082×10^{-5}). The highest value (0.212) was obtained in sunflower honey with 2.5% (w/w). This confirmed that the crystallisation rate could be accelerated by increasing the glucose content. Dettori *et al.* (2018) reported that the rate constant decreased with an increase in F/G ratio or reduced glucose, and the present data support this behaviour. When honey reaches the supersaturation level in the first phase, the nucleation rate is high. Based on this result, Avrami index or *n* values of sunflower honey (1.000 - 3.919) were higher than those of longan honey (1.000 - 1.529). This indicates that the crystal formation time of longan honey is longer than that of sunflower honey. Therefore, the Avrami model is a very appropriate model for describing the crystallisation behaviour from the intensity of absorbance values at 660 nm in sunflower and longan honey, under the condition of glucose addition. However, for further application with another honey type or any conditions, the relationship between the rate constant (*k*), Avrami index (*n*) and parameters of honey properties such as moisture content, initial concentration of sugar in honey, and water activity should be further considered.

Colour

Apart from smooth texture, colour of the final product in creamed honey is also a primary indicator of consumer acceptance. The honey colour changed during crystallisation process; a good creamed honey should be light yellow in colour which results from full fine crystals.

Figures 3A to 3F report the values of brightness (L*), hue angle (h°), and Whiteness Index (WI) of sunflower and longan honey samples from initial time until reaching full crystallisation under chill temperature storage. Before being induced by glucose powder, sunflower honey was clear to light yellow or golden-coloured, while longan honey presented a darker yellow colour. During the crystallisation process, L* and WI values increased in most of the samples, but rapidly changed in sunflower honey. The colour of longan honey slightly changed during storage (Figure 3D) and was similar to crystallisation behaviour.

Hue angle (h°) decreased until it reached full crystallisation, but it was not clearly changed in longan honey samples (Figure 3E). This is similar to the results of Dettori *et al.* (2018), who reported that h° of fast crystallisation honey sample showed low values because it contained 100% sunflower honey which was composed of high yellow and red components, which in turn gave a darker tint or lower h° as compared to slow and medium crystallisation samples. However, the colour was due to the appearance of glucose crystals reflecting the light; so, promoting the increase of brightness and a reduction of the yellow component resulted in a degree of the h° in all samples. The WI of sunflower honey was higher than longan honey, but showed similar characteristics with a longer time period (Figures 3C and 3F). Costa *et al.* (2015) reported that honey samples stored at 15°C gained higher values than stored at 25°C, and exponentially increased when the time increased. This increase in WI was due to the formation of crystals in the honey. Therefore, the colour and the crystallisation rate were involved in crystal formation during the crystallisation process.

Microstructure analysis

The crystals of honey samples in any treatment at fully crystallisation time are shown in Figures 3G and 3H. Both samples without glucose powder addition showed large, long, and sharp crystals occurrence (red and blue arrow). The crystal shape was pentagonal or hexagonal, and dispersed in single crystal. Mora-Escobedo *et al.* (2006) described the crystal appearance of Tajonal honey as a lattice with hexagonal and pentagonal crystals. The

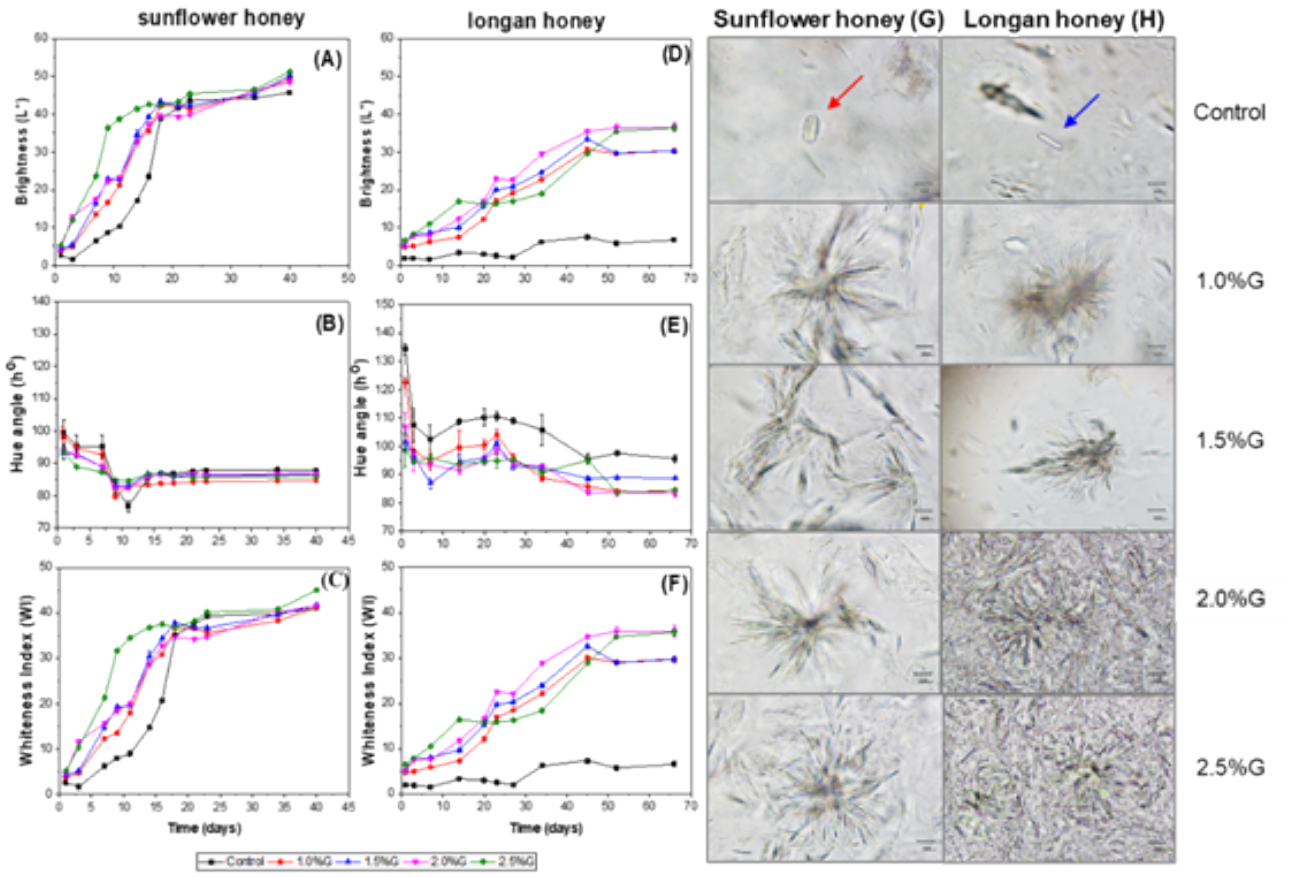

Figure 3. The L* value, hue angle (h°), and Whiteness Index (WI) of (A) - (C) sunflower honey, and (D) - (F) longan honey in different amounts of glucose powder addition during storage; and microscopic observation of honey crystals at the end of crystallisation for (G) sunflower honey, and (H) longan honey.

formation of glucose monohydrate crystals will impact the crystallisation patterns, in which small needle crystals were arranged in bouquets, as described by Dettori *et al.* (2018) and Lupano (1997) who reported the crystallisation process under cooling condition.

Larger bouquets of crystals were presented in longan honey with glucose addition, and less fine crystals were found (Figure 3H). This yielded coarse grain texture. However, many fine crystals were found in sunflower honey with glucose powder addition (Figure 3G). It also had a higher density than longan honey with increasing glucose addition. It was similar to the visible appearance of both samples. In terms of quantity, shape, and appearance, the varieties of crystals depended on the compositions and amount of glucose addition in the honey samples (Table 1 and 2). Karasu *et al.* (2015) mentioned that the texture of creamed honey was spreadable like butter, and was obtained due to the presence of a large number of very fine crystals in crystallised honey. Thus, sunflower honey may be suitable for the enhanced production of creamed honey.

Conclusion

The absorbance value at 660 nm is a good method to monitor the honey crystallisation kinetics, and the Avrami equation is a suitable model to explain the crystallisation behaviour with sunflower and longan honey when induced with glucose addition. Glucose addition impacted the crystallisation rate, colour, and crystal shape, which resulted from crystal formation during crystallization. Microstructural analysis of the small crystals found that crystals were arranged in bouquets, but were not different between treatments. Further studies should be conducted on other factors such as ultrasonic treatment and mixing temperature to control the crystallisation of creamed honey product. Storage tests in other conditions are also required.

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