International Food Research Journal 31(2): 352 - 367 (April 2024)
Journal homepage: *http://www.ifrj.upm.edu.my*

Characterisation of vacuum dried honey-sugar powder as affected by drying temperature and sugar carrier ratio for further application in chocolate

Rusli, A. A., *Mohamad, N. J., Mahmood, A. and Ibrahim, N. H.

Faculty of Fisheries and Food Science, Universiti Malaysia Terengganu, 21030 Kuala Nerus, Terengganu, Malaysia

Article history

Received:
8 May 2023
Received in revised form:
6 September 2023
Accepted:
4 January 2024

Keywords

honey powder, vacuum drying, hydroxymethylfurfural, diastase number, healthy chocolate

Abstract

The present work aimed to characterise oven-dried honey-sugar powders at vacuum temperatures (30 and 40°C) with dextrose, maltodextrin, and sucrose as the sugar carrier (incorporated at 50, 60, and 70%) for application in chocolate. Honey has high water content, and therefore, its direct use in chocolate will negatively affect the rheological properties of chocolate. Dextrose can produce a honey powder with the least heat degradation, hence maintaining the good health functionality of honey. Maltodextrin is the most common carrier used in honey-powder processing, while sucrose is the most common ingredient used in chocolate. The physical appearance and other physicochemical properties such as moisture content, hygroscopicity, glass transition temperature, hydroxymethylfurfural (HMF) content, and diastase activity were assessed for characterisation. Results showed that the honey-sugar powder that was dried at 40°C with 70% sugar carrier exhibited better properties in terms of physical appearance, moisture content, and hygroscopicity, with the HMF content and diastase activity not significantly affected by both factors. Therefore, honey-sugar powders dried at 40°C with 70% sugar carrier incorporation could have further application in chocolate. The impact of honey-sugar powder on chocolate rheological properties and their functionality in promoting health benefits is crucial for their potential application in the future.

DOI
https://doi.org/10.47836/ifrj.31.2.07

Introduction

Honey is an aromatic, sweet, and valuable natural product produced by honeybees. It contains a substantial proportion of low molecular weight (Mw) sugars, predominantly glucose (23 - 38%) and fructose (32 - 44%) (Mutlu *et al*., 2020), which contribute to its relatively low glass transition temperature (T_g). Honey offers several health benefits such as antioxidant, antidiabetic, anticancer, antibacterial, and wound healing (Moniruzzaman *et al*., 2013; Dan *et al*., 2018). In Malaysia, Tualang honey (TH) is the most favoured variety. TH distinguishes itself with a darker brown hue due to its heightened phenolic content compared to Gelam, Borneo, Pineapple, Acacia, and Kelulut honey (Khalil *et al*., 2011; Kishore *et al*., 2011; Moniruzzaman *et al*., 2013). The phenolic compounds found in TH encompass gallic, benzoic, syringic, *p*-coumaric, *trans*-cinnamic, and caffeic acids. TH also boasts flavonoid compounds like catechin, luteolin, naringenin, kaempferol, and apigenin (Khalil *et al*., 2011).

Chocolate is also known to possess various beneficial effects due to its substantial polyphenol content derived from cocoa. These effects encompass cancer risk prevention, cardiovascular disease reduction, and diabetes prevention (Godočiková *et al*., 2017). However, the elevated sugar content in chocolate can potentially lead to elevated blood sugar levels, and subsequently increase the risk of cardiovascular diseases, high blood pressure, and diabetes (Fernández, 2020). Numerous studies have advocated for the integration of sugar alternatives in chocolate formulations such as stevia, palm sugar, coconut sugar, inulin, polydextrose, maltodextrin, and sugar alcohols (Aidoo *et al*., 2014; Saputro *et al*., 2017; Aguilar-Villa *et al*., 2020; Ali *et al*., 2021). Yet,

*Corresponding author.
Email: niezaju@umt.edu.my

limited studies have been conducted on the substitution of sugar with honey powder in chocolate manufacturing.

Efforts to transform honey into powdered form for food applications have begun over the past three decades. Honey powders offer numerous advantages over liquid honey, including improved handling, extended shelf life, enhanced stability, and ease of incorporation into various food formulations (Nurhadi *et al*., 2012; Cuevas-Glory *et al*., 2017). Liquid honey has high water content, and therefore can detrimentally affect the rheological properties of chocolate and its processing (Afoakwa *et al*., 2007; Beckett, 2009). Most studies have focused on spray-drying honey powder due to its higher production rate, making it suitable for commercialisation (Shi *et al*., 2013; Samborska and Czelejewska, 2014; Suhag *et al*., 2018). However, alternative methods might be preferable for small-scale production, or when lower drying temperatures are desired. Freeze-drying, for instance, produces honey powder with elevated hygroscopicity, leading to a sticky and rubbery texture (Nurhadi *et al*., 2012). Meanwhile, vacuum-dried honey-maltodextrin powder at 60°C was found to exhibit increased HMF content and decreased diastase number (DN) (Nurhadi *et al*., 2012).

The issue of stickiness due to honey's low T_g can be resolved by incorporating a material with a higher T_g as a drying aid (Bhandari and Howes, 1999). Arabic gum and maltodextrin are commonly used for making honey powder. Arabic gum's T_g is approximately 126 to 194.5°C (Mothé and Rao, 2000; Nurhadi *et al*., 2012). However, excess Arabic gum (over 30%) can increase viscosity (Montenegro *et al*., 2012), and subsequently affect the hardness of foods that contain it. Maltodextrin, produced through starch hydrolysis, is characterised by its dextrose equivalent (DE), which indicates the quantity of reducing sugar present. Lower DE (5 - 10) maltodextrin has a T_g of 160 to 205.5°C, while higher DE (20 - 36) maltodextrin has a lower T_g, of 100 to 141°C (Hebbar *et al*., 2008; Nurhadi *et al*., 2012; Bhandari and Roos, 2016). Dextrose (glucose monohydrate) has also been utilised in honey drying; it is patented by Poltoratsky (2012), and has the least effect on DN. Sucrose is not a drying aid agent, which could potentially lead to stickiness during the drying process (Bhandari and Howes, 1999). However, sucrose is an important ingredient in chocolate which contains up to 70% (Afoakwa *et al*., 2007).

Findings on honey powder characteristics will guide the selection of honey powders for potential application in chocolate production in future research. To achieve its acceptable physical properties, particularly stability at room temperature, the proportion of drying agent in honey powder production should not fall below 50% of the solids (Nurhadi *et al*., 2012; Shi *et al*., 2013). Following this recommendation, the effects of vacuum drying temperature and sugar carrier ratio were examined, specifically the moisture content, hygroscopic rate, moisture sorption isotherm, glass transition temperature, solid-state structure, HMF content, and diastase number.

Materials and methods

Materials

Tualang honey was obtained from Hutan Hujan Tasik Kenyir, Kuala Terengganu, supplied by Koperasi Pemungut Madu Lebah Terengganu Berhad. Maltodextrin (dextrose equivalent, DE 10) was from Sim Supplies Sdn. Bhd. (Penang, Malaysia), dextrose from Eugene Chemical Sdn. Bhd. (Penang, Malaysia), and castor sugar (sucrose) from a local supermarket. Lithium chloride (LiCl), potassium acetate (CH_3COOK), magnesium chloride ($MgCl_2$), potassium carbonate (K_2CO_3), sodium nitrite ($NaNO_2$), sodium chloride (NaCl), 5-hydroxymethylfurfural, acetonitrile, and standards of fructose, glucose, maltose, and sucrose were from Sigma Chemical Co. (St. Louise, MO. U.S.A). HPLC-grade methanol (MeOH) was from Merck (Darmstadt, Germany). Phadebas tablets were from Magle AB (Lund, Sweden).

Preparation of honey-sugar powder

The formulations of TH powder are given in Table 1. The mixture was homogenised for 15 min at 4,000 *g* with a homogeniser (Ultra-turrax® T25 Digital Disperser, IKA, China). Then, 10 g of the mixture was weighed and spread on a Petri dish, and dried in a vacuum oven (Binder United States) at either 30 or 40°C under 25 in Hg for 6 h. Extending the drying time to 8 h showed no further reduction in moisture content. The dried samples were then ground into powder using mortar and pestle, and stored in a desiccator comprising P_2O_5 for further analysis.

Table 1. Formulations of honey-sugar powder.

Sugar carrier	Sugar carrier:Honey (w/w)		
	50:50	60:40	70:30
Dextrose	H/D 50	H/D 60	H/D 70
Maltodextrin	H/M 50	H/M 60	H/M 70
Sucrose	H/S 50	H/S 60	H/S 70

H: honey; D: dextrose; M: maltodextrin; and S: sucrose.

Determination of moisture content

The moisture content of honey-sugar powder was determined following the Association of Analytical Chemists (AOAC, 2000). Approximately, 3 g of honey-sugar powder was weighed into a pre-dried pan, and dried in a vacuum oven at 70°C for 6 h. The samples were prepared in triplicate, and data were recorded as mean ± standard deviation. The moisture content of honey-sugar powder was determined using Eq. 1:

$$Moisture = \frac{w1-w2}{w1}\ x\ 100 \quad \text{(Eq. 1)}$$

where, w1 = weight of the sample before (g), and w2 = weight of the sample after (g).

Determination of hygroscopicity rate

Approximately 1 g of honey-sugar powder was placed in a plastic vial, and stored in a desiccator containing a saturated NaCl solution to get 76% relative humidity (RH) at room temperature (Nurhadi *et al*., 2012). Samples were weighed every hour for 6 h.

Determination of moisture sorption isotherm

Moisture sorption isotherm was done by storing a sample of dried honey-sugar powder samples (1 g) in a desiccator, each containing different saturated salt solutions. The salt solutions used were LiCl, CH_3COOK, $MgCl_2$, K_2CO_3, $NaNO_2$, and NaCl for different water activities of 0.11, 0.23, 0.33, 0.44, 0.66, and 0.76, respectively. The weight of samples was monitored every 24 h for 14 days for a_w of 0.11, 0.23, 0.33, and 0.44, and 21 days for a_w of 0.66 and 0.76. After that, the samples were dried to determine their moisture content in a vacuum oven at 70°C for 6 h. The moisture sorption isotherm (MSI) curve was generated based on the data of water content corresponding to the different water activities. The GAB equation was used as a model of MSI (Nurhadi and Roos, 2016b) as shown in Eq. 2:

$$X = \frac{X_m C K a_w}{(1-K a_w)(1+C-1)K a_w} \quad \text{(Eq. 2)}$$

where, X = moisture content of the material on a dry basis, X_m = monolayer of water (kg of water/kg of dry matter), a_w = water activity, and C and K = constants.

Determination of X-ray diffraction pattern

XRD pattern analysis was conducted to determine the solid-state structure of dried honey-sugar powder, following the method outlined by Nurhadi *et al*. (2018). The honey-sugar powder was pressed into a rectangle sample holder (25 mm), and levelled with a blade. The analysis was carried out using an X-ray powder diffractometer (Rigaku MiniFlex, United States) at room temperature. The honey samples were scanned with 2θ between 10° to 80°, and the sugar carriers were scanned between 20° to 80°. The XRD pattern was observed using MiniFlex software installed in the equipment.

Determination of glass transition temperature

The T_g of dried honey-sugar powder was determined using DSC Q20 (TA instrument, Germany). This method was described by Nurhadi and Roos (2016a) with slight modifications. Honey sample (5 mg) was weighed in a 100 µL aluminium pan, and for reference, an empty pan was used. Sample was inserted at 25°C. Temperature scanning was first cooling down to -60°C at 20°C/min before going up to 200°C at 10°C/min. Heat flow calibration was performed using indium. Peaks were integrated using the TA-Universal Software analyser to determine the enthalpy (ΔH) and transition glass (T_g).

Determination of hydroxymethylfurfural

The HMF concentrations were determined using the services of a laboratory (Honey Quality Lab, Universiti Malaysia Terengganu) following the method outlined by International Honey Commission. About 10 g of each sample was diluted in 50 mL of distilled water, filtered through a 0.45 µm

nylon membrane filter, and about 20 µL samples were injected into an HPLC system (Shimadzu SPD-M20A, Kyoto, Japan) equipped with a Photodiode Array Detector (PDA). The HPLC column used was a Shim-pack GIST-HP C_{18} (150 × 4.6 mm, 5 µm) fitted with a guard cartridge packed with the same stationary phase (Shimadzu, Japan). An isocratic mobile phase consisting of 90% water and 10% methanol was used at a 1.0 mL/min flow rate. All solvents were of HPLC-grade. The identification wavelength ranged from 200 to 450 nm, with a specific monitoring at 285 nm. The quantification of HMF concentration was performed by comparing the peak area of the sample to that of standard 5-(hydroxymethyl) furan-2-carbaldehyde solutions (HMF). The results were expressed in mg/kg.

Determination of diastase activity

The diastase activity of dried honey-sugar powder was determined using the Phadebas method by International Honey Commission. A tablet of an insoluble blue-dyed, cross-linked starch was used as the substrate for the degradation reaction. About 13.6 g of sodium acetate trihydrate were dissolved in water to create an acetate buffer (0.1 M, pH 5.2). Glacial acetic acid was used to adjust the solution's pH to 5.2. The solution was then diluted with distilled water to a total volume of 1 L, and stored in a glass jar. About 1 g of sample was weighed and placed in a 100 mL volumetric flask which was then filled to the volume with the acetate buffer. About 5.0 mL of the sample was transferred to a new test tube, and incubated in a water bath at 40°C. A blank was prepared concurrently by adding 5.0 mL of the acetate buffer solution, which underwent the same treatment as the sample solution. After 10 min, a Phadebas tablet was introduced into both solutions, stirred for approximately 10 s, and returned to the 40°C water bath. Following a 30 min incubation, 1 mL of 0.5 M NaOH solution was added to stop the enzyme reaction. The solutions were stirred again and filtered. The absorbance at 620 nm was then measured using distilled water as the reference. The diastase activity was expressed as diastase number (DN), and calculated using Eq. 3:

$$DN = 35.2 \times \Delta A620 - 0.46 \quad \text{(Eq. 3)}$$

The powder was composed of 30, 40, or 50% of honey. After calculating the DN basic value, the DN for pure honey was determined by dividing 1 g of honey by the corresponding concentration of honey in the powder, and then multiplying it by the calculated DN. Considering moisture content, the impact of moisture was factored in using the moisture percentage of 1 g of honey. Subsequently, the calculation of dry matter involved subtracting the effect of moisture from the initial 1 g of honey. The DN on a dry basis was determined based on Göthe units/g of raw honey in honey powder.

Statistical analysis

All analyses were carried out in triplicate, and data were reported as mean ± standard deviation. Since data analysis using Two-way ANOVA showed no interaction between sugar carrier ratio and drying temperature, all samples were analysed using One-way ANOVA to compare means between samples. A comparison of the two temperatures was carried out using a *t*-test. Significant differences between samples were analysed using Tukey HSD (Honestly Significant Difference) multiple comparisons test at a 95% significance level. The software used for this statistical analysis was IBM SPSS Statistics 23.

Results and discussion

Physicochemical properties of Tualang honey

The raw TH recorded a moisture content of 22.1 g/100 g, which was comparable to the TH reported in other studies (in the range between 17.95 - 26.51%) (Chua *et al*., 2012; 2014; Zae *et al*., 2020). The moisture of raw honey should be less than 20% as outlined in the standard of Codex Alimentarius (2001). However, honey in tropical countries could reach up to 22% (Dan *et al*., 2018) due to the typical humid climate with high annual rainfall volume (Jaafar *et al*., 2012). The high moisture content as well as a significant amount of low molecular weight sugars (fructose and glucose) cause honey to have low glass transition temperature (T_g). In the present work, the T_g value of the raw TH was -59.45°C, comparable to Perhutani's honey at -47.5°C (Nurhadi *et al*., 2012), multi-floral Polish honey at -50.7°C (Bhandari and Roos, 2016), Spanish honey at -34.6°C to -47.15°C (Gómez-Díaz *et al*., 2012), and Indian nectar honey at -33.6°C to -51.1°C (Bhandari and Roos, 2016). The T_g value may vary based on water content and low Mw sugar composition in honey (Nurhadi and Roos, 2016a).

To ensure the raw TH is of good quality, an assessment of HMF and DN was carried out. HMF is

an indicator of the purity of honey, with high concentrations denoting excessive heating, poor storage conditions, and old honey (Mouhoubi-Tafinine *et al*., 2018). DN on the other hand is used as an indicator for honey freshness (Jaafar *et al*., 2012). High DN shows high freshness, while low DN shows over-heated, aged, and poorly stored honey samples (Chua *et al*., 2014). The HMF content and DN of the TH were 0.6 mg/kg. The raw TH used was considered fresh as the HMF value was below 10 mg/kg (Thrasyvoulou *et al*., 2018). The value obtained was slightly lower than the TH of other studies from Terengganu (0.75 mg/kg) and Kedah (1.7 mg/kg) (Jaafar *et al*., 2012; Fatima *et al*., 2016). The variations in HMF levels may be attributed to variations in flower sources, and their chemical properties such as reduced sugars, pH, total acidity, and mineral content; as well as external factors like temperature, heating time, storage condition, and the use of metallic containers (Mouhoubi-Tafinine *et al*., 2018). The DN value of the TH was 4.2 Göthe units, and therefore, complied with the Codex Alimentarius (2001) which stipulates that the DN value should not be less than 8. However, a value greater than 3 is considered acceptable for honey with low enzyme activity, particularly if its HMF value is lower than 15 mg/kg. The DN of TH in other studies were between 1.37 - 5.48 Göthe units (Jaafar *et al*., 2012; Chua *et al*., 2014). The high moisture of Malaysian honey can enhance the fermentation of yeasts, which release acid as the by-product. As a result, the pH is reduced, leading to a shorter half-life of the enzyme, and lowered DN values (Zulpa *et al*., 2021).

Physicochemical properties of honey-sugar powder

Physical appearance and moisture content of honey-sugar powder

The samples' physical appearances were observed and presented in Figures 1a, 1b, and 1c. As indicated, the honey-sugar powder dried at 40°C showed better powder appearance when contrasted with the samples dried at 30°C, which appeared lumpy and moist. An exception was noted for the H/S samples, whose textures were lumpy, wet, and sticky, regardless of both factors.

The appearance of honey-sugar powder is highly related to its moisture content (Table 2). However, the honey-dextrose (H/D) powder exhibited no significant differences in moisture content across all samples, while still displaying an improved powder appearance at 40°C in comparison to 30°C. A decreasing trend in moisture content was observed with increasing dextrose content, with minimal variation due to temperature increase. The drying process of the dextrose solution, *i.e*. the transitioning from dextrose hydrate to anhydrase dextrose, occurred at 50°C (Jackson and Silsbee, 1922), thus explaining the high moisture content in the H/D sample compared to other honey-sugar powders. Nevertheless, the moisture content of the H/D mixture remained generally comparable to the patented honey-dextrose mixture product, which was produced at temperatures ranging from 35 - 45°C (2 - 8%, w/w) (Poltoratsky, 2012).

The honey-maltodextrin (H/M) powder showed a decreasing trend as the maltodextrin increased. This trend was also observed with temperature variations, but only for maltodextrin concentrations of 50 and 60%. The H/M with 70% maltodextrin showed no differences as the temperature increased, indicating the pronounced influence of maltodextrin concentration due to a reduction in the availability of free water. A higher temperature is suggested, particularly for a high sugar concentration to increase the driving force for moisture evaporation (Islam *et al*., 2015). The moisture content obtained for H/M in the present work surpassed those of vacuum-dried honey-maltodextrin (50:50, w/w) sample dried at 60°C, as reported by Nurhadi *et al*. (2012). Nevertheless, the spray-dried honey-maltodextrin values in other studies ranged from 2.3 to 6.9% (Nurhadi *et al*., 2012; Cuevas-Glory *et al*., 2017; Suhag *et al*., 2018).

The H/S samples exhibited a texture having stickiness and lumpiness, although their moisture content was considerably lower than those of other samples. The stickiness in the H/S samples could be attributed to the cohesion-adhesion behaviour of sucrose particles. Cohesion manifested as stickiness between particles, while adhesion took place between particles and the wall surface (Muzaffar and Kumar, 2015). The H/S samples displayed particle sizes that were notably large and lumpy, primarily due to interactions between particles. This phenomenon led to the formation of inter-particle bridges, as discussed by Samborska and Czelejewska (2014) and Juarez-Enriquez *et al*. (2017). Water in the H/S samples was suspected of forming immobile liquid bridges, which create a strong binding force between the particles.

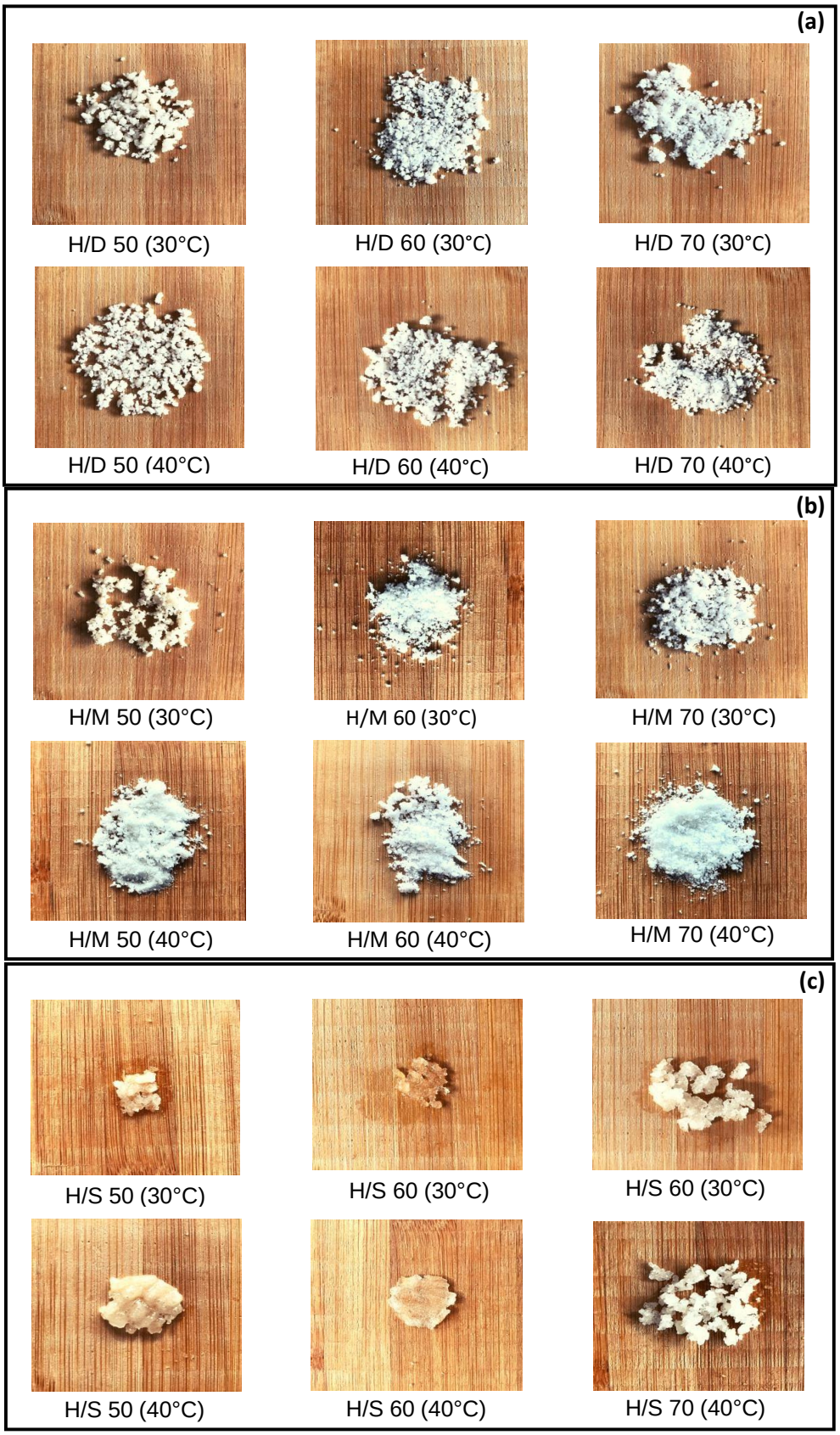


Figure 1. Physical appearances of honey-sugar powder **(a)** H/D, **(b)** H/M, and **(c)** H/S at different percentages (50, 60, and 70%) and temperatures (30 and 40°C). H: honey; D: dextrose; M: maltodextrin; and S: sucrose.

Table 2. Physicochemical properties (moisture content, hygroscopicity rate, hydroxymethylfurfural, and diastase number) of honey powder as affected by vacuum drying temperature and sugar carrier ratio.

Sample		Moisture content (g/100 g), wet basis		Hygroscopic rate (mg/h)		HMF (mg/kg)		Diastase number (Göthe units/g of raw honey in honey powder)	
		30°C	**40°C**	**30°C**	**40°C**	**30°C**	**40°C**	**30°C**	**40°C**
H/D	50%	7.8 ± 0.7^{Aa}	7.7 ± 0.2^{Aa}	9.4 ± 1.7^{Aa}	7.0 ± 1.3^{Aa}	4.2 ± 0.04^{Ab}	4.3 ± 0.52^{Ab}	2.7 ± 0.1^{Ab}	2.6 ± 0.0^{Aa}
	60%	7.7 ± 0.3^{Aa}	7.6 ± 0.1^{Aa}	7.0 ± 1.2^{Aab}	5.9 ± 1.0^{Aa}	5.3 ± 0.05^{Ba}	6.3 ± 0.57^{Aa}	3.0 ± 0.2^{Aab}	2.9 ± 0.4^{Aa}
	70%	7.6 ± 0.4^{Aa}	7.5 ± 0.1^{Aa}	5.6 ± 1.6^{Ab}	5.1 ± 1.5^{Aa}	5.4 ± 0.05^{Ba}	7.2 ± 0.06^{Aa}	3.3 ± 0.2^{Aa}	3.4 ± 0.4^{Aa}
H/M	50%	5.3 ± 0.2^{Aa}	4.3 ± 0.1^{Ba}	11.4 ± 1.7^{Aa}	8.2 ± 1.3^{Aa}	1.5 ± 0.01^{Bb}	3.4 ± 0.03^{Aa}	3.2 ± 0.1^{Ab}	3.2 ± 0.0^{Ab}
	60%	4.4 ± 0.1^{Ab}	4.1 ± 0.1^{Bab}	8.8 ± 1.2^{Aa}	7.1 ± 1.2^{Aa}	1.5 ± 0.07^{Bb}	4.1 ± 0.04^{Aa}	3.6 ± 0.4^{Aab}	3.5 ± 0.2^{Aab}
	70%	3.8 ± 0.3^{Ac}	3.8 ± 0.2^{Ab}	9.2 ± 1.6^{Aa}	7.5 ± 1.5^{Aa}	2.1 ± 0.02^{Ba}	4.1 ± 0.47^{Aa}	3.9 ± 0.1^{Aa}	3.6 ± 0.2^{Aa}
H/S	50%	4.9 ± 0.4^{Aa}	4.2 ± 0.2^{Aa}	5.9 ± 0.9^{Aa}	5.5 ± 0.9^{Aa}	4.9 ± 0.04^{Aa}	4.4 ± 0.53^{Aa}	4.0 ± 0.7^{Aa}	3.7 ± 0.2^{Aa}
	60%	4.7 ± 0.9^{Aa}	3.3 ± 0.3^{Ab}	6.1 ± 1.0^{Aa}	3.7 ± 0.4^{Bb}	4.3 ± 0.23^{Aa}	4.3 ± 0.04^{Aa}	3.9 ± 0.0^{Aa}	3.6 ± 0.3^{Aa}
	70%	4.4 ± 0.5^{Aa}	2.5 ± 0.1^{Bc}	5.1 ± 1.3^{Aa}	4.2 ± 0.2^{Aab}	4.4 ± 0.51^{Aa}	4.5 ± 0.51^{Aa}	3.7 ± 0.5^{Aa}	3.5 ± 0.2^{Aa}

Values are mean ± standard deviations of triplicates ($n = 3$). Means followed by different lowercase superscripts within the same column for each separated type of sugar carrier are significantly different ($p < 0.05$). Means followed by different uppercase superscripts within the same row for each separated type of sugar are significantly different ($p < 0.05$). H: honey; D: dextrose; M: maltodextrin; and S: sucrose.

Consequently, during the drying process for moisture content analysis, only a small fraction of water was able to evaporate, resulting in lower moisture content value, while concurrently presenting an undesired lumpy texture. The issue of stickiness has also been observed in high-sugar fruits such as bayberry and orange powder (Goula and Adamopoulos, 2010; Fang and Bhandari, 2012).

Hygroscopicity rate

Hygroscopicity is a crucial aspect of food powder quality as higher hygroscopicity is considered undesirable. The hygroscopic rates of the honey-sugar powders are shown in Table 2. Across all the honey-sugar powders, hygroscopicity rates fell within the range of 4.2 to 11.4 mg/h. These rates exhibited no significant differences with increases in both sugar content and temperature. Nevertheless, a decreasing trend was seen as the sugar concentration increased. Honey powder and coconut sap were noted to possess hygroscopicity rates of 14.4 and 8.86 mg/h, respectively (Nurhadi *et al*., 2012; 2018).

Moisture sorption isotherm

The sorption isotherm curves for H/D (Figure 2a), H/M (Figure 2b), and H/S (Figure 2c) showed fitting to the GAB model. The figures depict how the equilibrium moisture content increased in correspondence with higher water activities, aligning with the behaviour observed in other high-sugar foods that also showed water sorption capacity.

As tabulated in Table 3, the sorption isotherms of honey powders were classified as Type II, with the calculated values of K and C from the GAB model falling within the range $0.24 \leq kGAB \leq 1$ and $5.6 \leq CGAB \leq \infty$, respectively. This fell in line with the characteristics of a Type II isotherm, as noted by Lewicki (1997) and Suhag *et al*. (2018). Raw honey is typically characterised by a Type III isotherm, indicative of high-sugar foods (Mutlu *et al*., 2020). However, the inclusion of sugar as a carrier resulted in a transition to a Type II sorption isotherm, as discussed by Mutlu *et al*. (2020). This shift from Type III to Type II sorption isotherms has also been observed in the case of apple juice and pure pumpkin when added with inulin and egg albumin/methylcellulose, respectively (Jakubczyk *et al*., 2010; Stępień *et al*., 2022). Notably, other honey powders produced through vacuum and spray drying also exhibited The Type II sorption curves (Nurhadi and Roos, 2016b; Suhag *et al*., 2018; Mutlu *et al*., 2020).

The monolayer water content (X_m) serves as an indicator of food stability, with lower X_m values indicating an extended product's shelf life during storage. The X_m of honey powder values are shown in Table 3. As the quantity of sugar carrier increased, X_m decreased, with no discernible impact due to an increase in temperature (to 40°C) for all samples. This suggested that the honey-sugar powder containing 70% sugar carrier might have a longer shelf life than the sample with 50% due to the lower X_m value. The X_m values obtained for all samples were in line with reported values for honey powder in other studies, where carriers such as maltodextrin, Arabic gum, and whey protein isolate were used, resulting in X_m ranging from 3.78 to 7.9 g/100 g (Samborska *et al*., 2015; Nurhadi and Roos, 2016b; Mutlu *et al*., 2020). Comparable ranges were observed for other powders such as passion fruit powder (6.17 - 6.38 g/100 g), blackberry powder (7.1 - 8.7 g/100 g), and raspberry powder (7.4 - 10.9 g/100 g). These variations could be attributed to differences in chemical composition, leading to discrepancies in the GAB values and the hydrophilic/hydrophobic sites adsorbed at the interface (Syamaladevi *et al*., 2009; Pedro *et al*., 2010; Ferrari *et al*., 2012).

Glass transition temperature

T_g is a crucial parameter for explaining issues like stickiness during the drying process (Bhandari and Howes, 1999). In the present work, T_g values were determined for dextrose, maltodextrin, and sucrose, as well as for honey-sugar powders. However, due to limitations in the DSC instruments, the T_g values were not detected for dextrose and sucrose. The T_g value of maltodextrin was measured at 81.12°C, which was lower than the values reported for similar maltodextrins with comparable DE in previous studies (148.46 - 160°C) (Hebbar *et al*., 2008; Shi *et al*., 2013). Variations in the T_g value of maltodextrin can be attributed to differences in the physicochemical properties of starches from various botanical origins (Pycia *et al*., 2016). The T_g of dextrose was noted to be within the range of 31 to 38°C, while that of other works fell between 52 to 67°C (Hebbar *et al*., 2008; Bhandari and Roos, 2016).

Table 4 shows that T_g values for H/S samples were not detectable. Generally, the T_g values for all honey-sugar powders were higher than the T_g of raw

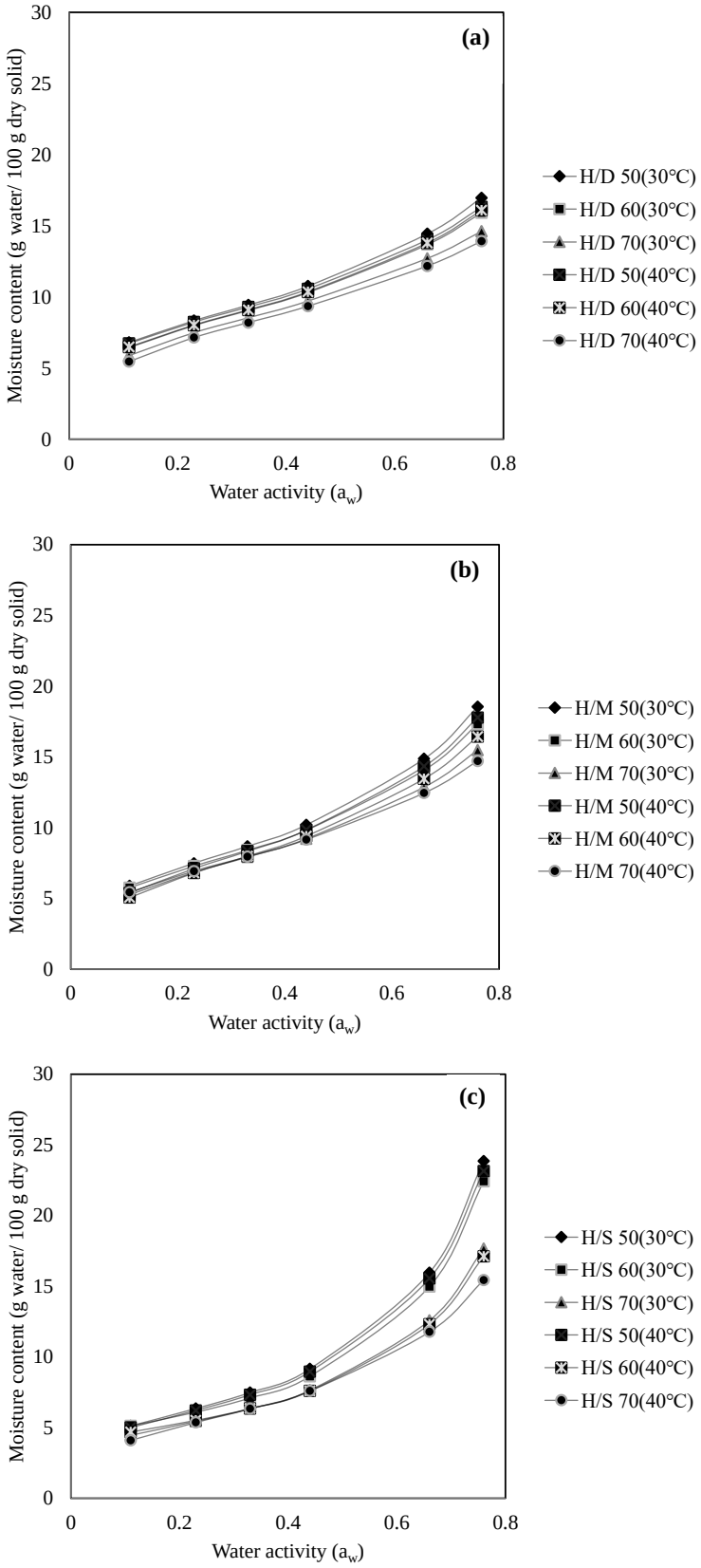


Figure 2. Moisture sorption isotherms of **(a)** H/D, **(b)** H/M, and **(c)** H/S at different percentages (50, 60, and 70%) and temperatures (30 and 40°C). H: honey; D: dextrose; M: maltodextrin; and S: sucrose.

Table 3. Estimated GAB model parameters for vacuum dried honey-sugar powder.

Sample		GAB Model							
		30°C				40°C			
		X_m	C	K	R^2	X_m	C	K	R^2
H/D	50%	7.7	53.4	0.7	0.99	7.6	53.5	0.9	0.98
	60%	7.5	46.3	0.9	0.99	7.5	47.1	0.7	0.98
	70%	7.4	36.0	0.7	0.99	7.4	29.3	0.7	0.99
H/M	50%	6.8	37.4	0.8	0.99	6.8	26.9	0.8	0.95
	60%	6.7	39.3	0.8	0.99	6.7	23.7	0.8	0.98
	70%	6.6	30.7	0.8	0.99	6.5	35.9	0.7	0.99
H/S	50%	5.1	64.6	1.0	0.95	5.1	61.8	1.0	0.90
	60%	4.7	23.3	1.0	0.98	4.4	23.8	1.0	0.98
	70%	4.4	25.8	1.0	0.95	4.3	27.4	1.0	0.93

H: honey; D: dextrose; M: maltodextrin; and S: sucrose.

Table 4. Glass transition temperatures of honey-sugar powder.

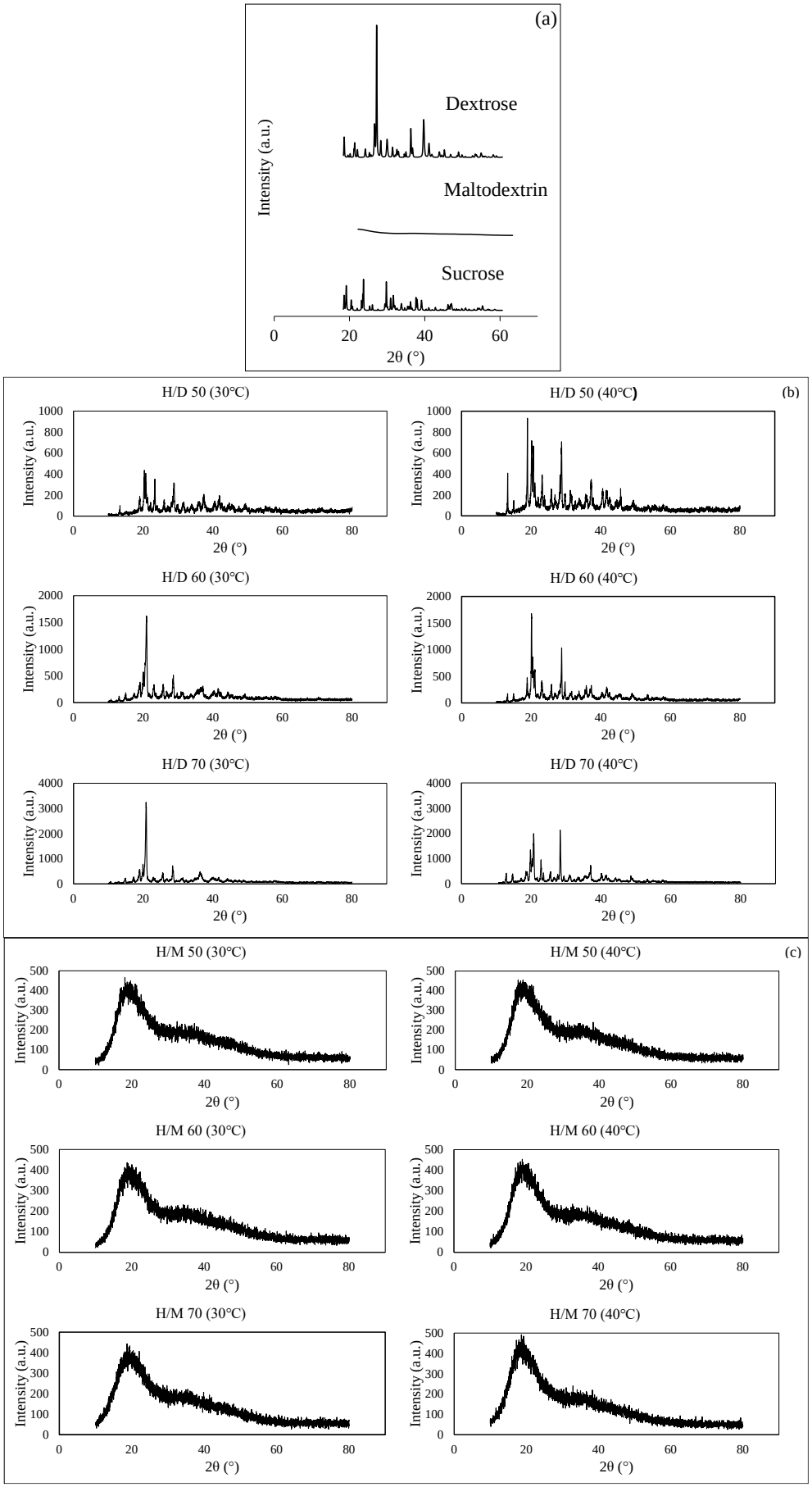
Sample		Glass transition temperature (°C)	
		30°C	40°C
H/D	50%	-22.48	-22.43
	60%	-20.86	-17.84
	70%	-20.46	-17.26
H/M	50%	62.57	62.71
	60%	68.35	70.09
	70%	73.81	76.36
H/S	50%	ND	ND
	60%	ND	ND
	70%	ND	ND

H: honey; D: dextrose; M: maltodextrin; and S: sucrose.

TH, ranging from -22.48 to 76.36°C, in contrast to the -59.45°C of raw honey. The T_g for H/D samples exhibited the highest value at -17.26°C, showing that they were likely in a rubbery state at room temperature as the value was significantly lower than at room temperature (Bhandari and Howes, 1999; Kaletunç, 2009). This state permits particle mobility, and can lead to structural changes like sticking, collapse, agglomeration, crystallisation, and more (Bhandari and Howes, 1999; Ahmed *et al*., 2017). Conversely, H/M samples exhibited higher T_g values, ranging from 62.57 to 76.36°C, indicating that they were in a glassy state due to their values exceeding ambient temperature. The glassy state is advantageous for powder drying as it leads to free-flowing properties (Ahmed *et al*., 2017). These T_g values surpass the value obtained for vacuum-dried honey powder (honey: maltodextrin, 56:44, w/w) of 25.99°C proposed by Osés *et al*. (2020). The lower value in that study might be attributed to the use of maltodextrin with a DE of 20. Maltodextrin with a higher DE value tends to correspond to a lower T_g value (Hebbar *et al*., 2008; Bhandari and Roos, 2016). Maltodextrin with a DE 10 is preferable due to its reduced degree of hydrolysation and fewer hydrophilic groups, resulting in lower hygroscopicity compared to maltodextrin with a higher DE value (Pycia *et al*., 2016).

XRD pattern of honey-sugar powder

Mixing honey with sugar carriers was expected to affect the solid-state structure of honey-sugar powder. Dextrose and sucrose exhibited a crystalline state structure, while maltodextrin showed an amorphous state structure (Figure 3a). The solid-state structures of the honey-sugar powder samples containing dextrose, maltodextrin, and sucrose are illustrated in Figures 3b, 3c, and 3d, respectively.


(a)
Dextrose
Maltodextrin
Sucrose
Intensity (a.u.)
2θ (°)
(b)
H/D 50 (30°C)
H/D 50 (40°C)
H/D 60 (30°C)
H/D 60 (40°C)
H/D 70 (30°C)
H/D 70 (40°C)
(c)
H/M 50 (30°C)
H/M 50 (40°C)
H/M 60 (30°C)
H/M 60 (40°C)
H/M 70 (30°C)
H/M 70 (40°C)

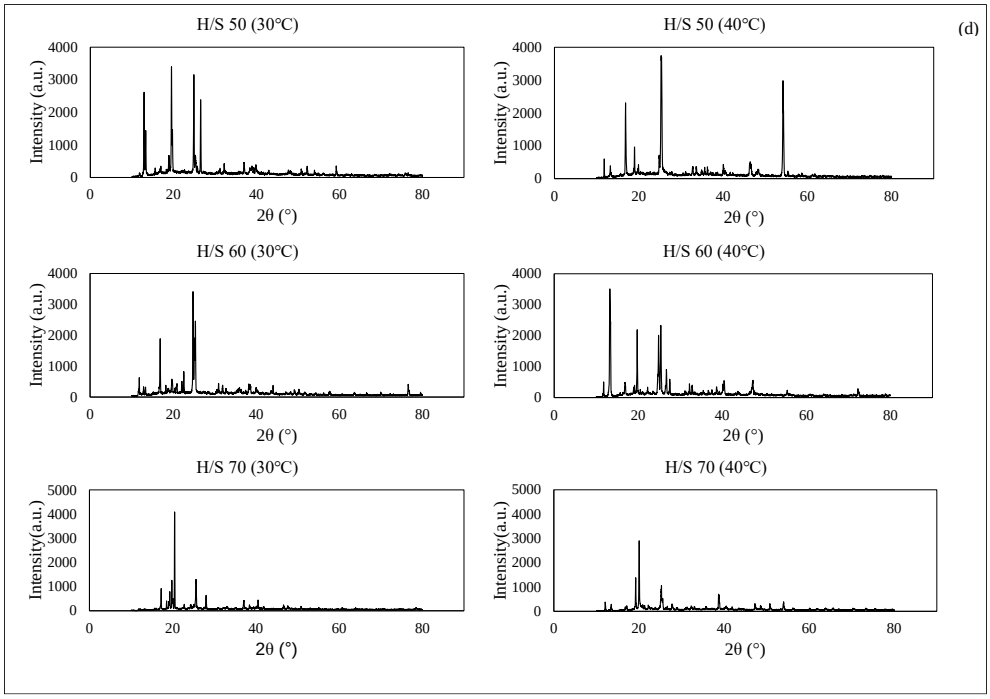


Figure 3. X-ray diffraction patterns of **(a)** pure sugar and honey-sugar powder **(b)** H/D, **(c)** H/M, and **(d)** H/S at the different percentages (50, 60, and 70%) and temperatures (30 and 40°C). H: honey; D: dextrose; M: maltodextrin; and S: sucrose.

Generally, the H/S powder showed the highest intensity compared to the other honey-sugar powders. This was likely due to the presence of numerous large lumpy structures in the H/S powder resulting from its sticky state. The H/D sample demonstrated increased intensity with higher concentrations, indicating an enhancement in the crystalline structure arising from the dextrose particles. Conversely, the H/M powder displayed the lowest intensity, with the pattern remaining consistent regardless of variations in concentration and drying temperature.

Hydroxymethylfurfural (HMF)

Generally, all the honey-sugar powder samples had HMF values higher than pure honey, thus indicating a significant effect from the sugar carriers and drying temperatures (Table 2). The HMF contents measured for all honey powders ranged from 1.5 to 7.2 mg/kg, which were within the acceptable range considering the maximum permissible statutory level set by the Malaysian Standard of 30 mg/kg of honey (Zulpa *et al*., 2021). The high HMF value results from the degradation of sugars during drying (Cuevas-Glory *et al*., 2017).

Diastase activity

Table 2 shows the DN of honey-sugar powder samples as affected by either drying temperature or sugar carrier concentration. The DN values ranged between 2.6 to 4.0 Göthe units, indicating only a slight decrease compared to the raw honey's value. DN values of all honey-sugar powders were also observed to be unaffected by drying temperatures, suggesting that the processing conditions preserved the diastase enzyme activity in the honey-sugar powder samples. The drying temperature should not exceed 40°C, or may lead to enzyme deactivation (Ariandi and Khaerati, 2017). Increasing the sugar carrier to 70% increased the DN of H/D and H/M samples. The removal of water in honey-sugar powder was shown to enhance enzyme stability by restricting the freedom of movement of the protein molecules. This phenomenon inhibited conformational changes that could otherwise lead to activity deterioration, as elucidated by Samborska and Czelejewska (2014).

Conclusion

The honey produced at 40°C exhibited superior characteristics across all assessed parameters, particularly in formulations with the highest carrier incorporation. Conversely, the drying process at 30°C yielded unsatisfactory results. Notably, the H/M powder displayed the most favourable attributes in terms of moisture content, monolayer value, glass transition temperature, and overall physical appearance moisture. The moisture sorption isotherm

was also satisfactory for H/D and H/M samples, unlike the H/S sample which displayed a pronounced tendency to absorb water, particularly at the highest applied water activity of 0.8. This attribute led to noticeable large lumpy and sticky texture in the H/S sample. To better understand the effects of heat degradation for compounds like HMF and diastase number, it is thus recommended that experiments be conducted at higher drying temperatures, such as 50 and 60°C.

Honey-sugar powders that contained 70% carrier, and dried at 40°C were suggested for further application in chocolate production. Given the concerns of high sugar content (up to 70%) in chocolate, the substitution of sugar with honey-sugar powder presents an opportunity to mitigate the associated health risks by enhancing the existing polyphenols in cocoa chocolate. The incorporation of H/D, H/M, and H/S into chocolate warrants further investigation into its rheological properties, antioxidant capacity, and alpha-amylase inhibition. These analyses could discern the impacts of dextrose, maltodextrin, and sucrose. Potential applications in yoghurt and ice cream are also recommended for future research.

Acknowledgement

The manuscript is an original research work carried out at the Faculty of Fisheries and Food Science, Universiti Malaysia Terengganu, Malaysia. There is no copyright infringement in the figures or any part of the manuscript. The present work was financially supported by the Fundamental Research Grant Scheme (FRGS) awarded by the Ministry of Higher Education, Malaysia (grant no.: FRGS/1/2018/WAB01/UMT/03/5).

References

Afoakwa, E. O., Paterson, A. and Fowler, M. 2007. Factors influencing rheological and textural qualities in chocolate - A review. Trends in Food Science and Technology 18(6): 290-298.

Aguilar-Villa, C., Acosta-Otálvaro, E. V., Rodríguez-Sandoval, E. and Mazo-Rivas, J. C. 2020. Sucrose-free milk chocolate manufacture using bulking agents and a non-caloric sweetener. Food Science and Technology 40(1): 62-67.

Ahmed, J., Rahman, M. S. and Roos, Y. H. 2017. Glass transition and phase transitions in food and biological materials. United States: John Wiley and Sons.

Aidoo, R. P., Afoakwa, E. O. and Dewettinck, K. 2014. Optimization of inulin and polydextrose mixtures as sucrose replacers during sugar-free chocolate manufacture - Rheological, microstructure and physical quality characteristics. Journal of Food Engineering 126: 35-42.

Ali, A. M., Shekib, L. A., Elshimy, N. M. and Sharara, M. S. 2021. Producing and quality attributes of low calories dark chocolate using different intense sweeteners and wheat fiber isolate. American Journal of Food Science and Technology 9(1): 1-7.

Ariandi, and Khaerati. 2017. Test the enzyme activity of diastase, hydroxymethylfurfural (HMF), reducing sugar content, and water content in Battang forest honey. Prosiding Seminar Hasil Penelitian 2017: 1-4.

Association of Official Analytical Chemists (AOAC). 2000. Official method of analysis of AOAC International. 17th ed. United States: AOAC.

Beckett, S. T. 2009. Chocolate manufacture. In Talbot, G. (ed). Science and Technology of Enrobed and Filled Chocolate, Confectionery and Bakery Products, p. 11-28. United States: Woodhead Publishing.

Bhandari, B. and Roos, Y. H. 2016. Non-equilibrium states and glass transitions in foods. United States: Woodhead Publishing.

Bhandari, B. R. and Howes, T. 1999. Implication of glass transition for the drying and stability of dried foods. Journal of Food Engineering 40(1): 71-79.

Chua, L. S., Abdul-Rahaman, N. L., Sarmidi, M. R. and Aziz, R. 2012. Multi- elemental composition and physical properties of honey samples from Malaysia. Food Chemistry 135(3): 880-887.

Chua, L. S., Adnan, N. A., Abdul-Rahaman, N. L. and Sarmidi, M. R. 2014. Effect of thermal treatment on the biochemical composition of tropical honey samples. International Food Research Journal 21(2): 773- 778.

Codex Alimentarius. 2001. Codex Stan 12-1981 - Revised Codex standard for honey. Rome: FAO.

Cuevas-Glory, L. F., Pino, J. A., Sosa-Moguel, O., Sauri-Duch, E. and Bringas-Lantigua, M. 2017. Optimization of the spray-drying process

for developing stingless bee honey powder. International Journal of Food Engineering 13(1): 20160217.

Dan, P. N. S. M., Omar, S. and Ismail, W. I. W. 2018. Physicochemical analysis of several natural Malaysian honeys and adulterated honey. IOP Conference Series - Materials Science and Engineering 440(1): 012049.

Fang, Z. and Bhandari, B. 2012. Comparing the efficiency of protein and maltodextrin on spray drying of bayberry juice. Food Research International 48(2): 478-483.

Fatima, I. J., Mohd Hilmi, A. B., Salwani, I. and Lavaniya, M. 2016. Physicochemical characteristics of Malaysian stingless bee honey from *Trigona* species. International Medical Journal Malaysia 17(1): 187-191.

Fernández, A. R. G. 2020. Physicochemical properties and sensory acceptability of sugar free chocolate formulations added with fish oil and probiotics. Mexico: University of Monterrey, MSc thesis.

Ferrari, C. C., Germer, S. P. M., Alvim, I. D., Vissotto, F. Z. and de Aguirre, J. M. 2012. Influence of carrier agents on the physicochemical properties of blackberry powder produced by spray drying. International Journal of Food Science and Technology 47(6): 1237-1245.

Godočiková, L., Ivanišová, E. and Kačániová, M. 2017. The influence of fortification of dark chocolate with sea buckthorn and mulberry on the content of biologically active substances. Advanced Research in Life Sciences 1(1): 26-31.

Gómez-Díaz, D., Navaza, J. M. and Quintáns-Riveiro, L. C. 2012. Physicochemical characterization of Galician honeys. International Journal of Food Properties 15(2): 292-300.

Goula, A. M. and Adamopoulos, K. G. 2010. A new technique for spray drying orange juice concentrate. Innovative Food Science and Emerging Technologies 11(2): 342-351.

Hebbar, U. H., Rastogi, N. K. and Subramanian, R. 2008. Properties of dried and intermediate moisture honey products: A review. International Journal of Food Properties 11(4): 804-819.

Islam, M. Z., Kitamura, Y., Yamano, Y. and Kitamura, M. 2015. Effect of vacuum spray drying on the physicochemical properties, water sorption and glass transition phenomenon of orange juice powder. Journal of Food Engineering 169: 131-140.

Jaafar, M. H. M., Hamid, K. A., Anuar, N., Zohdi, R. M. and Effendi, T. J. B. 2012. Physicochemical properties and pharmacokinetic profiles of selected Malaysian honey. In 2012 IEEE Symposium on Business, Engineering and Industrial Applications, p. 140-145. Bandung, Indonesia.

Jackson, R. F. and Silsbee, C. G. 1922. The solubility of dextrose in water. United States: US Government Printing Office.

Jakubczyk, E., Ostrowska-Ligeza, E. and Gondek, E. 2010. Moisture sorption characteristics and glass transition temperature of apple puree powder. International Journal of Food Science and Technology 45(12): 2515-2523.

Juarez-Enriquez, E., Olivas, G. I., Zamudio-Flores, P. B., Ortega-Rivas, E., Perez-Vega, S. and Sepulveda, D. R. 2017. Effect of water content on the flowability of hygroscopic powders. Journal of Food Engineering 205: 12-17.

Kaletunç, G. 2009. Calorimetry in food processing: Analysis and design of food systems. United States: John Wiley and Sons.

Khalil, M. I., Alam, N., Moniruzzaman, M., Sulaiman, S. A. and Gan, S. H. 2011. Phenolic acid composition and antioxidant properties of Malaysian honeys. Journal of Food Science 76(6): 921-928.

Kishore, R. K., Halim, A. S., Syazana, M. S. N. and Sirajudeen, K. N. S. 2011. Tualang honey has higher phenolic content and greater radical scavenging activity compared with other honey sources. Nutrition Research 31(4): 322-325.

Lewicki, P. P. 1997. The applicability of the GAB model to food water sorption isotherms. International Journal of Food Science and Technology 32(6): 553-557.

Moniruzzaman, M., Khalil, I., Sulaiman, S. A. and Gan, S. H. 2013. Physicochemical and antioxidant properties of Malaysian honeys produced by. BMC Complementary and Alternative Medicine 13(43): 1-12.

Montenegro, M. A., Boiero, M. L., Valle, L. and Borsarelli, C. D. 2012. Gum Arabic: More than an edible emulsifier. Products and Applications of Biopolymers 51: 953-978.

Mothé, C. G. and Rao, M. A. 2000. Thermal behavior of gum Arabic in comparison with cashew gum. Thermochimica Acta 357-358: 9-13.

Mouhoubi-Tafinine, Z., Ouchemoukh, S., Bachir Bey, M., Louaileche, H. and Tamendjari, A. 2018. Effect of storage on hydroxymethylfurfural (HMF) and color of some Algerian honey. International Food Research Journal 25(3): 1044-1050.

Mutlu, C., Koç, A. and Erbaş, M. 2020. Some physical properties and adsorption isotherms of vacuum-dried honey powder with different carrier materials. LWT – Food Science and Technology 134: 110166.

Muzaffar, K. and Kumar, P. 2015. Parameter optimization for spray drying of tamarind pulp using response surface methodology. Powder Technology 279: 179-184.

Nurhadi, B. and Roos, Y. H. 2016a. Dynamic water sorption for the study of amorphous content of vacuum-dried honey powder. Powder Technology 301: 981-988.

Nurhadi, B. and Roos, Y. H. 2016b. Water sorption and water plasticization behavior of vacuum died honey. International Journal of Food Properties 19(6): 1370-1380.

Nurhadi, B., Andoyo, R., Mahani and Indiarto, R. 2012. Study the properties of honey powder produced from spray drying and vacuum drying method. International Food Research Journal 19(3): 907-912.

Nurhadi, B., Sukri, N., Sugandi, W. K., Widanti, A. P., Restiani, R., Noflianrini, Z., … and Herudiyanto, M. 2018. Comparison of crystallized coconut sugar produced by traditional method and amorphous coconut sugar formed by two drying methods: Vacuum drying and spray drying. International Journal of Food Properties 21(1): 2339-2354.

Osés, S. M., Cantero, L., Crespo, M., Puertas, G., González-Ceballos, L., Vallejos, S., … and Sancho, M. T. 2020. Attributes of ling-heather honey powder obtained by different methods with several carriers. LWT – Food Science and Technology 150: 112063.

Pedro, M. A. M., Telis-Romero, J. and Telis, V. R. N. 2010. Effect of drying method on the adsorption isotherms and isosteric heat of passion fruit pulp powder. Food Science and Technology 30: 993-1000.

Poltoratsky, V. B. 2012. U.S. Patent application no. 13/001,417. United States: United States Patent and Trademark Office.

Pycia, K., Juszczak, L., Gałkowska, D., Mariusz, W. and Jaworska, G. 2016. Maltodextrins from chemically modified starches. Part 2 - Selected physicochemical properties. Carbohydrate Polymers 146: 301-309.

Samborska, K. and Czelejewska, M. 2014. The influence of thermal treatment and spray drying on the physicochemical properties of Polish honeys. Journal of Food Processing and Preservation 38(1): 413-419.

Samborska, K., Langa, E. and Bakier, S. 2015. Changes in the physical properties of honey powder during storage. International Journal of Food Science and Technology 50(6): 1359-1365.

Saputro, A. D., Van de Walle, D., Aidoo, R. P., Mensah, M. A., Delbaere, C., De Clercq, N., … and Dewettinck, K. 2017. Quality attributes of dark chocolates formulated with palm sap-based sugar as nutritious and natural alternative sweetener. European Food Research and Technology 243(2): 177-191.

Shi, Q., Fang, Z. and Bhandari, B. 2013. Effect of addition of whey protein isolate on spray-drying behavior of honey with maltodextrin as a carrier material. Drying Technology 31(13-14): 1681-1692.

Stępień, A., Witczak, M. and Witczak, T. 2022. The thermal characteristics, sorption isotherms and state diagrams of the freeze-dried pumpkin-inulin powders. Molecules 27(7): 2225.

Suhag, Y., Nayik, G. A. and Nanda, V. 2018. Modelling of moisture sorption isotherms and glass transition temperature of spray-dried honey powder. Journal of Food Measurement and Characterization 12(4): 2553-2560.

Syamaladevi, R. M., Sablani, S. S., Tang, J., Powers, J. and Swanson, B. G. 2009. State diagram and water adsorption isotherm of raspberry (*Rubus idaeus*). Journal of Food Engineering 91(3): 460-467.

Thrasyvoulou, A., Tananaki, C., Goras, G., Karazafiris, E., Dimou, M., Liolios, V. and Gounari, S. 2018. Legislation of honey criteria and standards. Journal of Apicultural Research 57(1): 88-96.

Zae, T. K., Azlan, A., Sajak A. B., Hock, E. K., Nyuk, L. C., Md Noh, M. F. and Mustafa Khalid, N. 2020. Comparison of selected local honey with manuka honey based on their nutritional and antioxidant properties. Food Research 4: 203-215.

Zulpa, A. K., Mohd Isa, N. S., Ahmad, F. T., Islamiah, M. N. and Yusof, H. M. 2021. Quality characteristics, antioxidant and anticancer potential of stingless bee honey and honeybee honey from similar environmental conditions. IIUM Medical Journal Malaysia 20(4): 29-36.