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Influence of Anti-Caking Agent on the Water Sorption Isotherm and Flow-ability Properties of Vacuum Dried Honey Powder

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Highlights (For Review)

- Effect of anti-caking agent addition to amorphous honey powder on its static and dynamic water sorption
- Effect of anti-caking agent addition to amorphous honey powder on its powder flow ability properties
- Effect of anti-caking agent addition on inhibiting recrystallization of amorphous sugar in vacuum-dried honey powder

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Influence of Anti-Caking Agent on the Water Sorption Isotherm and Flow-ability Properties of Vacuum Dried Honey Powder

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ABSTRACT

Honey powder is a hygroscopic powder due to its composition and structure. The addition of anti-caking agent was aimed to increase the stability of honey powder. The present study aimed to study the influence of anti-caking agent on the water sorption isotherm and flow-ability properties of vacuum dried honey powder. Anti-caking agents, calcium silicate and calcium stearate, were added in honey powder. The addition of anti-caking agent influenced water sorption and flow-ability properties of honey powder. Calcium silicate addition seemed not to affect water sorption isotherm properties of honey powder, however, calcium stearate showed more inhibiting effect of recrystallization and collapse of structure of amorphous honey powder. Addition of anti-caking agent to honey powder increase flow-ability of honey powder and calcium stearate seemed to increase flow-ability better than calcium silicate.

***Key Words**: Honey powder, anti-caking, calcium silicate, calcium stearate, flow-ability, water sorption isotherm*

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1. Introduction

Honey powder is a hygroscopic powder due to its composition and structure. The two main sugars found in honey solids, fructose, and glucose, have very low glass transition temperatures making honey powder hygroscopic (Bhandari and Howes, 1999). Nurhadi and Roos (2016a) reported the production of honey powder by using vacuum drying technique and drying aid materials (filler), maltodextrin. Anhydrous honey powder glass transition temperature was reported over 30 to 43 °C depending on drying aid material type and composition (Nurhadi and Roos, 2016a). Critical water content of honey powder was also very low, less than 2 % (g water/100 g sample) (Nurhadi and Roos, 2016a). Honey powder might be in purely amorphous or partially amorphous structure depending on process used (Nurhadi and Roos, 2016b). When water is sorbed by honey powder during storage, water acts as a plasticizer, reduce glass transition temperature of honey powder to lower than ambient temperature, results in significant changes of macroscopic properties of honey powder, i.e. collapse which includes phenomena such as stickiness, caking and shrinkage (Roos, 1995; Bhandari and Howes, 1999). The rate of changes was dependent on the difference of ambient temperature to glass transition temperature ($T-T_g$), the higher the difference was the faster was the rate at which the macroscopic properties changed (Roos, 1995; Foster, Bronlund, Paterson, 2006; Roos and Drusch, 2015). The change would reduce quality of honey powder and the powder is becoming difficult to handle and difficult to flow.

Method to keep honey powder stable during storage is by storing it at a temperature below its glass transition temperature or below its critical water activity (Ca_w). Additionally, anti-caking agent could be used to maintain stability of sugar-rich powder such as honey powder (Welti-Chanes et al., 2007). Anti-caking agent (or flow conditioner/free flowing agent) is the ingredient added at low quantity to increase stability and maintain free flowing properties of host material (Barbosa-Canovas et al., 2005). Anti-caking agent mechanism is different for amorphous and crystalline materials (Msagati, 2013). The mechanism of anti-caking agent might be by competing for water with host material, acting as a water barrier, eliminating surface friction and inhibiting crystal growth (Barbosa-Canovas et al., 2005 and Lipasek et al., 2012). Anti-caking agent is classified as natural (such as kaolin, talc, and bentonite/silicate material) or synthetic when

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manufactured from raw materials such as silicon dioxide (Msagati, 2013). Lipasek et al (2012) reported the use of silicon dioxide, calcium silicate and calcium stearate up to 2 % as anti-caking agent in host powder of crystalline material. The addition of anti-caking agent might affect water sorption isotherm and flowing properties of host powder. The addition of anti-caking agent into crystalline material did not change the shape of water sorption isotherm and some anti-caking agent showed delaying deliquescence of crystalline material (Lipasek, Taylor and Mauer, 2011 and Lipasek et al., 2012).

Powder flow-ability is more complex than fluid (Xanthakis et al., 2015). Powder flow-ability is affected by material properties and stress history of material during processing (Koynov, Glasser, Muzzio, 2015). Powder flow-ability is affected by internal factor such as particle size and distribution and external factor such as air humidity and temperature (Barbosa-Canovas et al., 2005). Powder flow-ability is one of powder bulk properties (Barbosa-Canovas et al., 2005). Powder would flow if the load acting on the powder is bigger than the powder strength (or the bulk structure of powder failed and start moving) (Barbosa-Canovas et al., 2005). Barbosa-Canovas et al (2005) considered powder flow-ability as a failure phenomenon. Two main properties used to describe the flow-ability of powder are failure function and effective angle of internal fraction. Failure function correlates major principal consolidation stress (MPS) and unconfined failure strength (UFS) to deduce the value of flow index. The correlation between MPS and UFS is linear and the inverse slope of the best fit linear line equals to the flow index (Barbosa-Canovas et al., 2005, Koynov, Glasser and Muzzio, 2015). The slope describes the change of stress needed to induce the powder flow. The lower is the slope, the more free flowing properties of the powder exist. The effective angle of internal friction is a property that describes the inter particle interaction (Barbosa-Canovas et al., 2005) and powder with a low value of effective angle is considered as a free flowing powder. The current study was aimed to investigate the influence of anti-caking agents on water sorption and flow-ability properties of anhydrous vacuum-dried honey powder.

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2. Materials and Methods

2.1. Honey Powder

Honey was bought from local supermarket (Boyne valley Honey Brand) in Ireland. Deionized water was used from KB scientific (Cork, Ireland) and maltodextrin DE 10 (M100) was obtained from Grain processing corporation (IA, USA). Honey-maltodextrin solutions (total solid 43 % and ratio honey: maltodextrin = 6:4 and 4:6 (honey composition of 60 % and 40 % per total solid, respectively)) were prepared on petri dishes with perforated covers for drying in vacuum oven (WTB Binder, Germany) at temperature 60 °C for 3 hours. The mixture of honey, maltodextrin and water is named as HMW. HMW 4:6 and HMW 6:4 referred to honey powder with honey composition of 40 % and 60 % per total solid respectively. The dried samples were ground and stored in desiccator over P_2O_5 (Sigma-Aldrich) for further analysis

2.2. Mixing Honey Powder and Anti-Caking Agent

Anti-caking agents, calcium silicate and calcium stearate (Sigma-Aldrich), were used. Anti-caking agents in Erlenmeyer flask and honey powder were added gradually based on geometric mixing principle to obtain an even distribution. Each addition of honey powder was followed by mixing with spatula.

2.3. Water Sorption Isotherm

2.3.1. Static water sorption

Static water sorption was prepared by storing sample of dried honey powder ($\pm$ 1 gram) in desiccator over various saturated salt solutions. Seven different salt solutions were put in desiccator to adjust the relative humidity (RH). The salt solutions used were LiCl, CH_3COOK, $MgCl_2$, K_2CO_3, $Mg(NO_3)_2$, $NaNO_2$, and NaCl (Sigma Chemical Co., St. Louise, MO. U.S.A) to adjust RH value of 11 %, 23 %, 33 %, 43 %, 52 %, 65 % and 75 %, respectively (Roos, 1995). The weight change of samples was monitored regularly (every day) for three weeks or until steady weight was reached. The water content of samples from each desiccator were determined by drying in vacuum oven at 100 °C for 6 hours. The data of water content of honey powder and their

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corresponding water activity was used to make water sorption isotherm (WSI) curve. The Guggenheim-Anderson-deBoer (GAB) equation was used as a model of WSI.

$$X = \frac{X_m C K a_w}{(1 - K a_w)(1 + (C - 1) K a_w)} \qquad (1)$$

Where,

X = water content (g water/ g dry solid)

a_w = water activity

X_m = monolayer water content

X_m, C, and K are constants

2.3.2. Dynamic water sorption (Dynamic Dew point Isotherm/DDI)

Dried sample (about 500 mg) was used in DDI (Decagon Devices, Inc. Pulman WA, USA) measurements. The adsorption direction of dynamic water sorption was set up. The measurement were conditioned as follows, temperature of 25 °C, air flow of 300 ml/min and a_w range starting from 0.1 to 0.85. In DDI adsorption direction, the wet air pass over the dried sample and the sample would adsorb moisture from the air. If the initial water activity of dried sample higher than 0.1 a_w, the dry air would pass over the sample until its water activity less than 0.1, then adsorption direction would be started. The data of sample weight change and its corresponding water activity were recorded at certain interval time. The initial water content of dried sample was determined by reducing water activity below 0.1 a_w and the predicted water content then processed by DDI software (SorpTract Software, Decagon, Device Inc., Pulman WA, USA). The data collection continued until the final set water activity was reached. The data of sample weight changes were converted into data of sample water content. Finally the data of sample water content and its corresponding water activity would be used to draw dynamic water sorption curve.

2.4. Tapped Density

Honey powder sample was put in 25 ml measuring cylinder until fully covered the volume stated. The measuring cylinder with honey powder inside then was put in tapped density meter (J.

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Engelsmann AG, Germany) and tapping was done for 100 times. The end volume was then recorded. Hausner Ratio (H_R) was calculated as followed (Barbosa-Cannovas et al., 2005):

$$H_R = \frac{\rho_\infty}{\rho_o}$$

Where :

ρ_∞ = tapped density (bulk density of powder after tapping)

ρ_o = initial loose bulk density

2.5. Powder Flow ability

Flow ability of honey powder was measured by using Powder Flow tester (Brookfield, USA). Sample of honey powder was put in trough whose annular shape and flattened with inner catch tray. The trough has volume 38 cc. the initial weight of trough and the weight after sample addition were recorded. The vane lid was then moved down to cover the trough. The vane lid had diameter of 12.7 cm and curved profile. Standard flow test was set to run the experiment. The trough then rotated while powder consolidated/compressed. Five consolidation stresses were used. Five consolidation stresses used were 0.795, 1.607, 3.246, 6.559 and 13.252 kPa. As an example at consolidation stress of 0.795 kPa, the 3 stresses set points used were 0.795 KPa and the other two stresses lower than 0.795 kPa (0.265 and 0.536 kPa) (Fig. 1). The trough rotated until steady flow reached and then the shear stress and normal stress were recorded. Data of normal stress and shear stress were recorded for each consolidation stress. From Fig. 1, major principal consolidation stress (MPS) and unconfined failure strength (UFS) were determined for each consolidation stress with the use of powder flow pro. V1.2 Build 19 (Brookfield, USA). The best fit line of normal stress and shear stress was called as yield locus and Mohr's semi-circle was used to determine MPS and UFS (Barbosa-Cannovas et al., 2005 and Koynov, Glasser and Muzzio, 2015). UFS was determined with the use of Mohr's semi-circle which passing through origin point and tangent to the yield locus while MPS was determined with the use of Mohr's semi-circle which passing through pre shear consolidation stress (0.597 KPa) and tangent to the yield locus. MPS and UFS represented the condition for the critical state and critical failure respectively

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(Koynov, Glasser dan Muzzio, 2015). Effective yield locus was determined based on the line which passing through origin and tangent to bigger Mohr's semi-circle and the angle between x-axes and effective yield locus was called as effective angle of internal friction (Barbosa-Cannovas et al., 2005). The data between MPS and UFS for every consolidation stress was then correlated to determine flow factor index. Flow factor index was the inverse of slope of best fit linear line between MPS and UFS.

Based on the value of flow factor index (ff), flow ability of a powder can be classified as (Barbosa-Cannovas et al., 2005).

ff < 1 non flowing

1 < ff <2 very cohesive

2 < ff < 4 cohesive

4 < ff < 10 easy flowing

ff > 10 free flowing

Fig 1. Yield Locus, effective yield locus, and Mohr's stress semi-circle from a sample of honey powder (HMW 4:6) flow ability measurement by powder flow tester at 0.795 kPa consolidation stress.

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3. Results and Discussion

3.1. Water Sorption Isotherm

Water sorption isotherm of honey powder was done by using static/conventional and dynamic method (DDI method). Even though the data from dynamic water sorption could not be compared with data obtained from conventional method in calculating water sorption model (such as GAB and BET model) (Schmidt and Lee, 2012), the DDI data could be used to study the properties changes related to their corresponding water activities (Romani et al., 2016) such as critical water activity determination (Yuan, Carter and Schmidt, 2011; Carter and Schmidt, 2012), deliquescent point (Ghorab et al., 2014) and amorphous content (Nurhadi and Roos, 2016b). From Fig. 2, it can be seen that addition of anti-caking agent (2%) to host powder of honey powder did not change shape of WSI and the WSI were classified as type II of Brunaeur water sorption isotherm classification (Labuza and Altunakar, 2007). Nurhadi and Roos (2016a) also reported that honey powder had type II of Brunaeur water sorption isotherm classification. The result agreed with those reported by Lipasek, Taylor and Mauer (2011) that the anti-caking agent did not change the WSI shape of deliquescent material. As shown in Fig. 2, anti-caking agent addition to honey powder also did not change its corresponding monolayer water content.

Fig. 2. Static water sorption isotherm of honey powder and honey powder with addition of anti-caking agent and GAB parameter for each treatment.

Honey powder (maltodextrin ratio of 60 % per total solid) with calcium stearate addition (2%) had higher water sorption than other two treatments of honey powder over water activity (a_w) range of 0.44-0.76 (Fig. 2). As shown by Fig. 3a, the significant increase of water sorption of honey powder occurred above a critical water activity (around 0.35 a_w). The Ca_w of honey powder was higher than the Ca_w of honey powder (determined based on static water sorption) reported by Nurhadi and Roos (2016a). The different result was caused by the different water sorption method and the Ca_w obtained from dynamic WSI determination usually higher than the Ca_w value based on static WSI and DSC (differential scanning calorimeter) glass transition measurement (Carter and Schmidt, 2012). The critical value obtained by DDI method is much affected by the

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flow rate of air over the sample during the measurement. The higher is the air flow rate, the higher is the critical water activity obtained (Yuan, Carter and Schmidt, 2011). The Ca_w from normal method (DSC and WSI method) might be equal to the Ca_w obtained from DDI method by extrapolating the Ca_w at zero flow rate (Yuan, Carter and Schmidt, 2011). From Fig. 3.a. It could be seen that anti-caking addition seemed not to change the critical water activity of honey powder. The critical water activity was correlated with the significant increase of water sorption due to transition from glassy structure to rubbery structure of amorphous component (Yuan, Carter and Schmidt, 2011; Carter and Schmidt, 2012). After critical water activity surpassed, the macroscopic properties such as crystallization of amorphous components and caking increased significantly (Nurhadi and Roos, 2016b). As shown in Fig. 3a, there were differences in water sorption after 0.5 a_w between honey powder with calcium stearate and other two honey powders (honey powder and honey powder with calcium silicate). From DDI WSI result, the "equilibrium water content" for both calcium stearate and calcium silicate corresponded to 0.1-0.8 a_w, were of 0.2 % - 0.5% and 1.8 % - 6.0 %, respectively. The low water sorption of both calcium stearate and calcium silicate were also reported by Lipasek et al. (2012).

The clearer differences of water sorption behavior between honey powders are shown in Fig. 3b. Nurhadi and Roos (2016b) used first derivative of DDI WSI curve to show crystallization of amorphous components during water sorption which appeared as a peak and the area of peak correlated to the extent of crystallization occurred. It seemed that the crystallization of amorphous components of honey powder occurred, however, the inhibition of crystallization was apparent in honey powder with addition of calcium stearate. Honey powder without anti-caking agent and honey powder with calcium silicate had more complete peak than the honey powder with calcium stearate (Fig. 3b).

a.

b.

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Fig. 3. Dynamic water sorption of honey powder and honey powder with the addition of anti-caking agent (a), first derivative curve of dynamic water sorption of honey powder and honey powder with the addition of anti-caking agent (b).

From static WSI experiment, the collapsed structure of honey powder appeared in honey powder without anti-caking agent and honey powder with calcium silicate starting at 0.44 a_w while collapse did not occur in honey powder with calcium stearate (Fig. 4). However at higher a_w, honey powder with Calcium stearate addition also showed stickiness and lump phenomena (0.65 and 0.75 a_w) and need tapping the vial several time to move out the powder from the vial. Calcium stearate act as a barrier and due to its hydrophobic properties to repel water from ambient air to the host particle (honey powder) that might show some inhibition effect of crystallization of amorphous sugar (maintaining structural integrity) and collapse. Lipasek et al. (2012) explained the ability of calcium stearate as moisture barrier between particles that might interrupt bridging mechanism as an initial step of collapse occurred. Murrieta et al. (2011) stated the collapsed structure of milk powder was triggered by recrystallization of amorphous lactose in milk powder. The collapse phenomena occurred when Ca_w surpassed and caused physical and chemical changes such as non-enzymatic browning reaction (Roos, 1995, Roos and Drusch, 2015). Thus it could be shown that the addition of anti-caking agent (Calcium stearate) into honey powder gave some effect of inhibition of crystallization and collapse of amorphous structure of honey powder during water sorption experiment. That explained the phenomena occurring in static WSI of honey powder (Fig. 2) where honey powder with calcium stearate had higher water sorption than the other two honey powders due to recrystallization inhibition of amorphous sugar in honey powder. The addition of anti-caking agent, calcium silicate and calcium stearate at 1% in honey powder showed the same result as those with 2% (data not shown). The disadvantages of calcium stearate addition are related to its taste and solubility.

Fig. 4. Honey powder and honey powder with addition of anti-caking agent after reaching equilibrium at various water activity, from top to bottom correspond to honey powder, honey powder + 2 % calcium stearate and honey powder + 2 % calcium silicate, respectively.

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3.2. Flow-ability

In order to work optimally as anti-caking agent, there should be interaction or surface affinity between anti-caking agent and host powder (Barbosa-Canovas et al., 2005). The interaction is indicated by the increase of bulk density of host powder after addition of an anti-caking agent (Barbosa-Canovas et al., 2005). As shown in Table 1, the addition of anti-caking agent (calcium stearate and calcium silicate) increased bulk density from 0.59 to 0.64-0.66 (g/cm^3). The increase of bulk density showed the high surface affinity between anti-caking and host particle and if it is not, anti-caking agent could fill the void between host particles powder and no increase of bulk density may occur (Barbosa-Canovas et al., 2005).

Table 1. Parameters of honey powder obtained from powder flow tester and tapped density tester.

Flow-ability of honey powder with higher maltodextrin ratio was better than the lower ratio. As given in Table 1 and Fig. 5, the flow-ability index of honey powder increased from cohesive to easy flowing powder when the ratio of maltodextrin to honey increased from 40 % to 60 % (solid ratio). Honey powder with lower maltodextrin ratio had lower glass transition temperature (Nurhadi and Roos, 2016a) and tended to be more hygroscopic. The DDI critical water activity of honey powder increased (from around 0.3 a_w to 0.35 a_w) when the maltodextrin ratio to honey increased from 40 % to 60 % (per total solid) (Fig. 6). Stoklosa et al. (2012) stated that flow-ability of powder was influenced significantly by external conditions such as air relative humidity of storage. When water was adsorbed by honey powder, water acted as a plasticizer and reduced glass transition temperature of honey powder to below ambient temperature (Roos and Drusch, 2015) and honey powder became more sticky and difficult to flow.

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Fig. 5. The flow-ability characteristic of honey powder and honey powder with addition of anti-caking agent.

Fig. 6. Dynamic water sorption of honey powder at different ratio of maltodextrin DE 10

Instead of increasing the ratio of maltodextrin as a drying aid material, anti-caking agent might be used to improve the flow-ability of honey powder. As can be seen in Table 1 and Fig. 5, the addition of anti-caking agent 2 % (both calcium stearate and calcium silicate) could increase the flow-ability properties of honey powder from an easy flowing to a free flowing powder. Density properties were also used as an indication of powder flow-ability (Barbosa-Canovas et al., 2005 and Xanthakis, Ommen and Ahrne, 2015). Relative decrease of bulk density after compaction or tapping of powder was an indicator that the powder had good flow-ability properties (Barbosa-Canovas et al., 2005). From Table 1, based on the value of Hausner ratio (H_R), the honey powders (excluding honey powder with lower maltodextrin ratio, HMW 6:4) were classified as a medium flowing powder while HMW 6:4 was classified as a difficult flowing powder. The increasing trend of bulk density value after compaction in powder flow-ability tester of honey powder with anti-caking agent were smaller than that of honey powder with anti-caking agent and the result complied with the flow index results. However, the contrary result based on density measurement was found in honey powder with different maltodextrin ratio. The contrary result of flow-ability measurement based on density properties was also reported by Xanthakis, Ommen, and Ahrne (2015).

Flow-ability of powder was determined as a failure function in term of flow index as explained in Barbosa-Canovas et al (2005). In addition to flow index, the effective angel of internal friction can also be used to determine the flow-ability properties of honey powder. The lower the value of effective angle of internal friction, the more free flowing properties the powder has (Barbosa-Canovas et al., 2005). As shown in Fig. 7, honey powder with calcium stearate had the lowest effective angle of internal friction followed by both honey powder without anti-caking agent and honey powder with calcium silicate and honey powder with lower maltodextrin ratio (HMW 6:4). The effective angle of internal friction is a property that described

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inter-particle interaction of powder. The lower value of the angle indicated a smoother surface interaction between particles in the powder. Compared to calcium silicate, calcium stearate has bigger molecular size and had ability to elongate and narrow better than calcium silicate to give protection of the surface more extensively. Lipasek et al (2012) also reported calcium stearate as the best anti-caking agent in reducing effective angle of internal friction of crystalline sucrose.

Fig. 7. Effective angle of internal friction of honey powder and honey powder with addition of anti-caking agent.

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4. Conclusion

Anti-caking agent calcium silicate and calcium stearate were added into honey powder. The addition of anti-caking agent influenced water sorption and flow-ability properties of honey powder. Calcium silicate addition seemed not to affect water sorption isotherm properties of honey powder, however calcium stearate showed some inhibiting effect of recrystallization and collapse of structure of amorphous components of honey powder. Addition of anti-caking agent to honey powder increase flow-ability of honey powder and calcium stearate seemed to increase flow-ability better than calcium silicate. Addition of anti-caking agent increased flow-ability of honey powder from an easy flowing to a free flowing powder and calcium stearate showed significant reduction of internal friction between particles of honey powder that contributed the powder becoming more free flowing.

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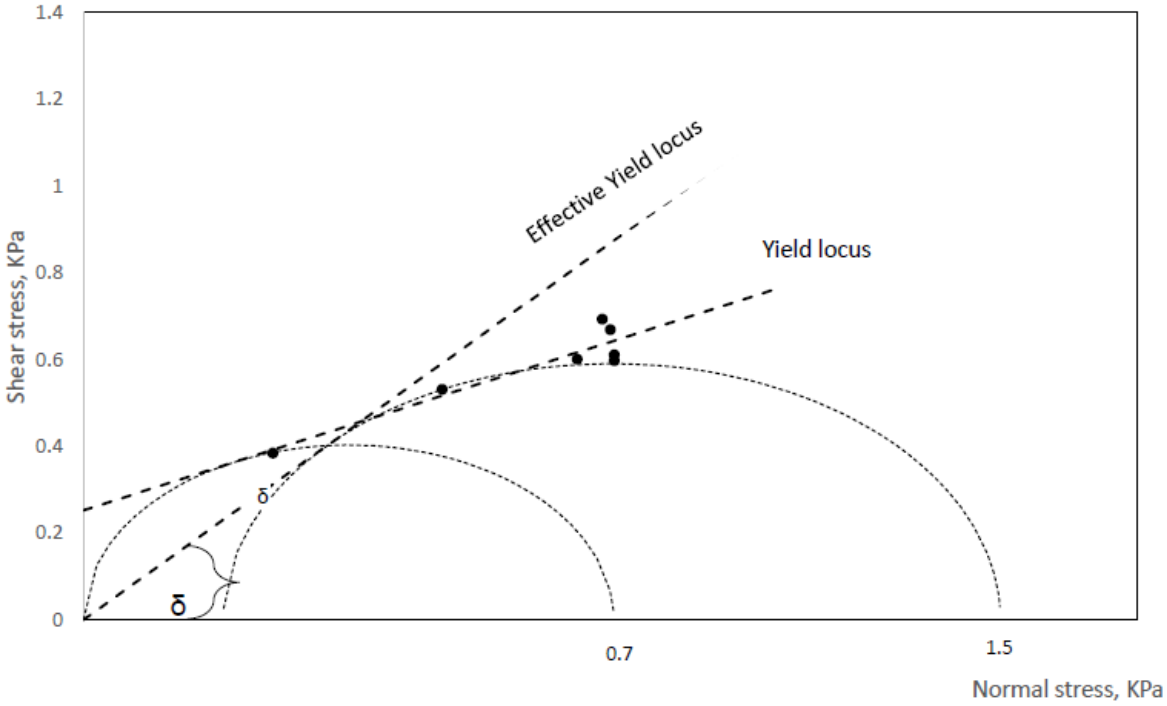


Fig 1. Yield Locus, effective yield locus, and Mohr's stress semi-circle from a sample of honey powder (HMW 4:6) flow ability measurement by powder flow tester at 0.795 kPa consolidation stress.

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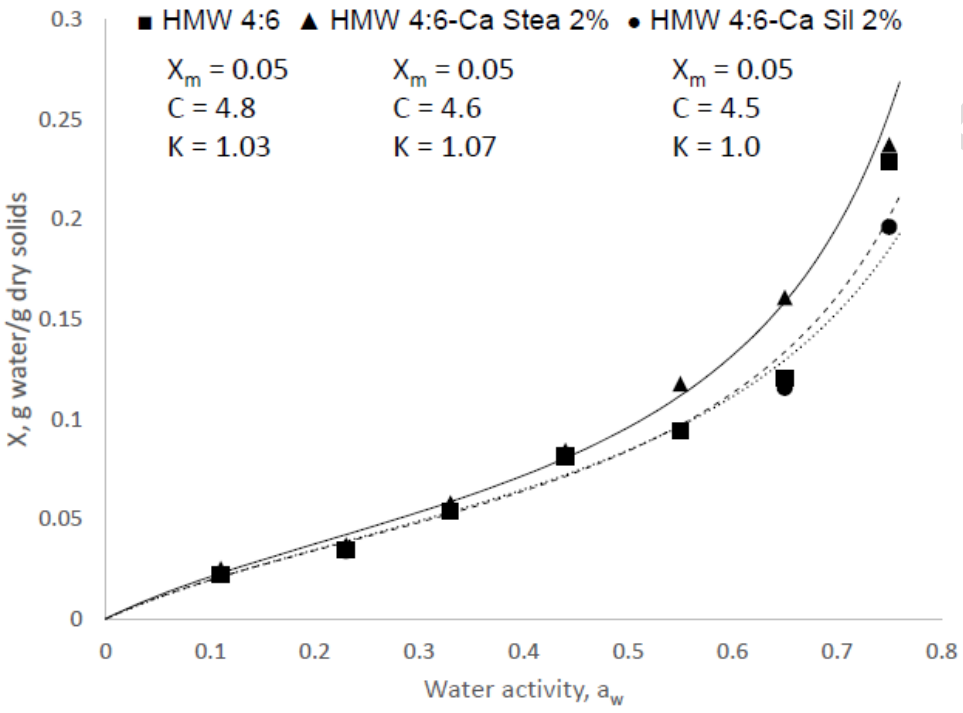
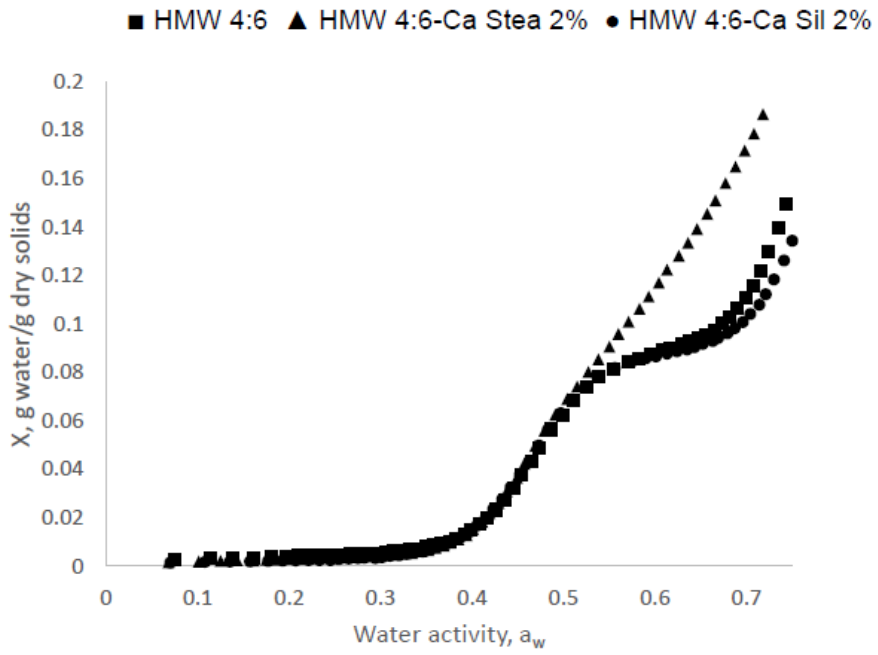
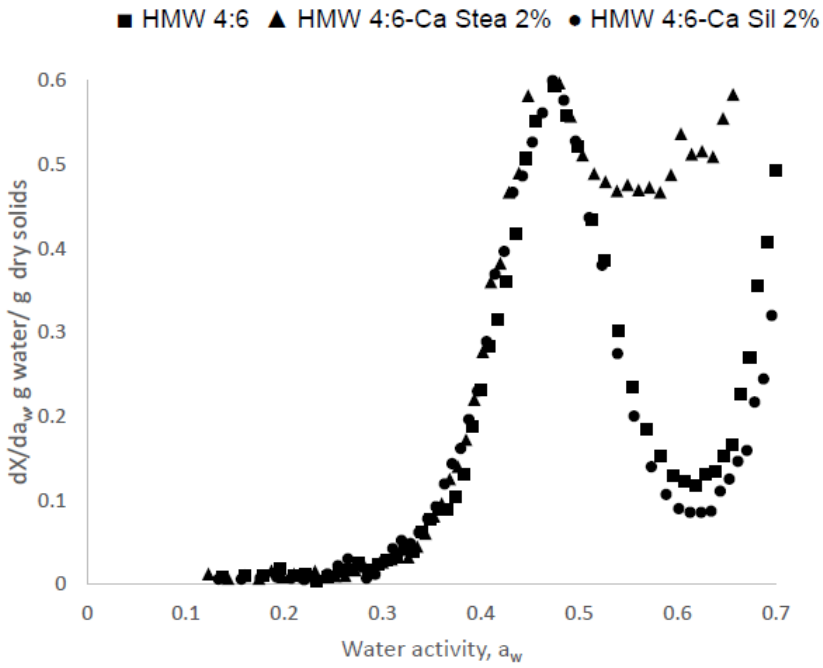


Fig. 2. Static water sorption isotherm of honey powder and honey powder with addition of anti-caking agent and GAB parameter for each treatment.

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a.



b.

Fig. 3. Dynamic water sorption of honey powder and honey powder with the addition of anti-caking agent (a), first derivative curve of dynamic water sorption of honey powder and honey powder with the addition of anti-caking agent (b).

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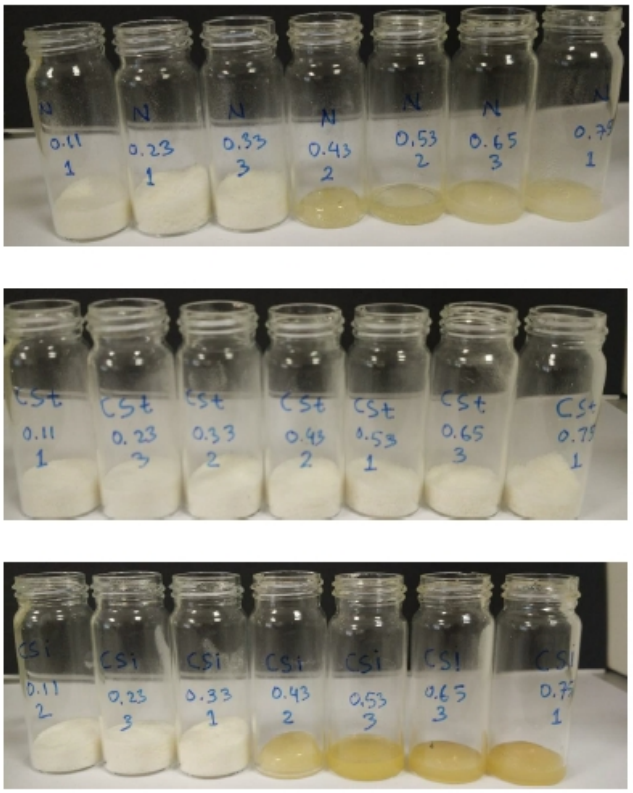


Fig. 4. Honey powder and honey powder with addition of anti-caking agent after reaching equilibrium at various water activity, from top to bottom correspond to honey powder, honey powder + 2 % calcium stearate and honey powder + 2 % calcium silicate, respectively

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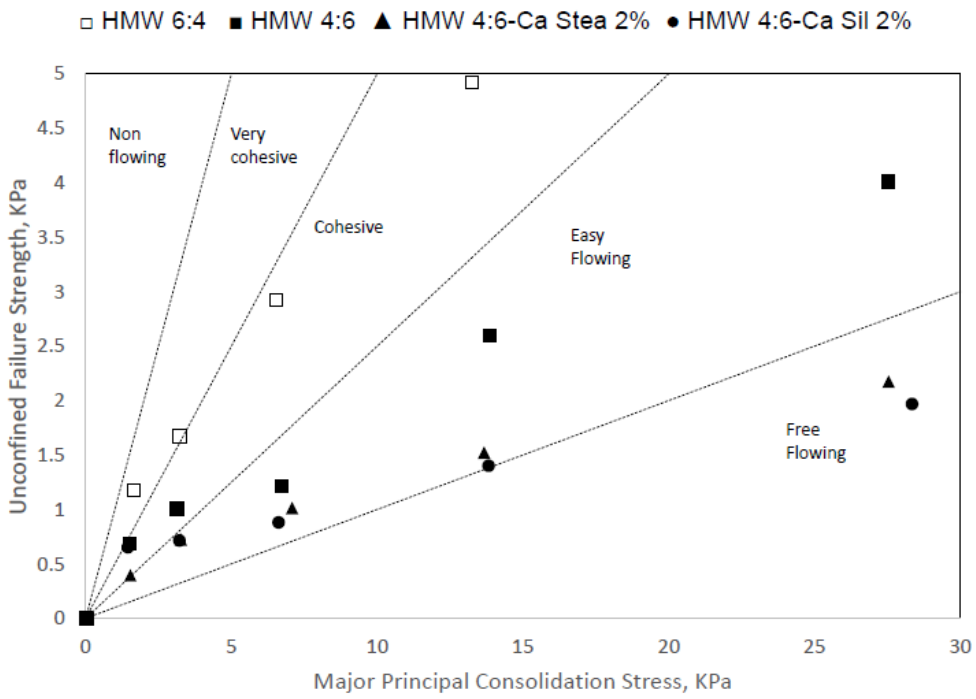


Fig. 5. The flow-ability characteristic of honey powder and honey powder with addition of anti-caking agent.

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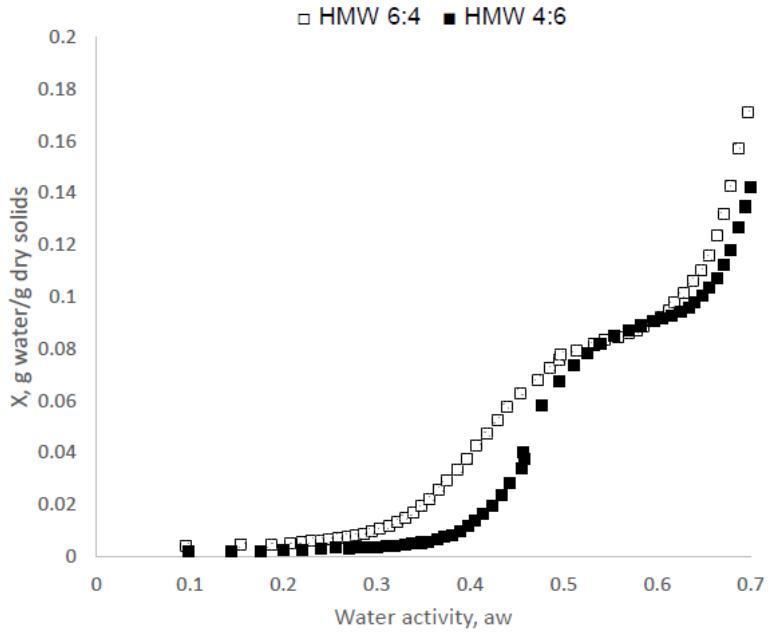


Fig. 6. Dynamic water sorption of honey powder at different ratio of maltodextrin DE 10

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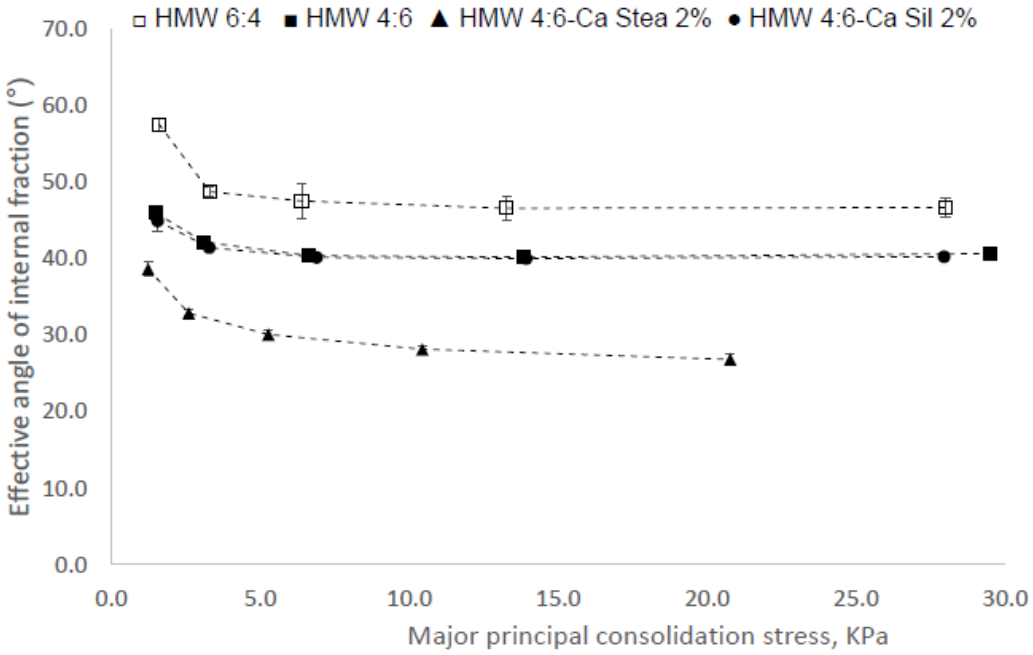


Fig. 7. Effective angle of internal friction of honey powder and honey powder with addition of anti-caking agent.

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Table 1. Parameters of honey powder obtained from powder flow tester and tapped density tester.

Treatment	Powder flow-ability tester				Tapped Density (g/cm^3)		
	Flow index, ff	Density (g/cm^3)					
		Initial	After Consolidation 6.5 kPa	Relative Increase	ρ_o	ρ_t	H_R
HMW 6:4	3.04 ± 0.26	0.49 ± 0.03	0.72 ± 0.02	1.49 ± 0.03	0.58 ± 0.01	0.75 ± 0.00	1.3 ± 0.03
HMW 4:6	7.64 ± 0.43	0.59 ± 0.01	0.91 ± 0.03	1.57 ± 0.06	0.67 ± 0.01	0.81 ± 0.01	1.21 ± 0.00
HMW 4:6-Ca Stea 2 %	11.91 ± 2.55	0.66 ± 0.03	1.05 ± 0.12	1.49 ± 0.11	0.70 ± 0.01	0.85 ± 0.02	1.22 ± 0.00
HMW 4:6-Ca Sil 2 %	13.44 ± 0.38	0.64 ± 0.01	0.90 ± 0.02	1.39 ± 0.03	0.70 ± 0.00	0.81 ± 0.00	1.16 ± 0.00

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