THE INVESTIGATION OF THE APPLICATION OF SKIM MILK AS A CARRIER AND DILUENT DURING HONEY SOLUTION SPRAY DRYING

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Summary. Maltodextrin and skimmed milk powder were used (separately) as carriers for the spray drying of multiflower honey. Main while, water and liquid skimmed milk were used as two different diluents for the dissolution of honey to facilitate spray drying. The application of skimmed milk improved the product's yield and enabled the increase of honey content in the received powders up to 70% of solids that cannot be observed when water is used a diluent and maltodextrin as a carrier. Moreover, milk used as a carrier as well as a diluent, had a positive effect on values of water content, water activity, density, cohesiveness and morphology, improving the overall quality of finished product comparing to variants where maltodextrin was used as a carrier and water as a diluent. Based on the obtained results, it was concluded that milk powder can be successfully used as an alternative to maltodextrin as a carrier in honey spray drying.

Key words: honey powders, multiflower honey, spray drying, carrier agents, skimmed milk

INTRODUCTION

Honey is one of the products that has a positive effect on health because of the presence of many important and valuable ingredients such as enzymes, acids, antioxidants and minerals. The valuable properties of this product have been widely used in pharma-

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cology, medicine, cosmetics and food industry over the years. However, due to its sticky consistency and crystallisation tendency, the possibilities of using this product in the food industry are limited. Spray drying is a solution to this issue that enables to produce the honey in the form of a powder which eliminates the technical difficulties that are associated with its viscus nature like dosing, handling and storing [Samborska 2019].

Honey is characterised by high content of low molecular weight monosaccharides (glucose and fructose), which are challenging during spray drying due to their low glass transition temperature (T_o). At that temperature, honey sugars which exist in amorphous state changes into the rubbery state inside the drying chamber resulting in stickiness and inferior powder's yield and quality. This problem can be managed by using a carrier material that have a high molecular weight and high T_{e} value, which compensates for the low T_{σ} of honey's monosaccharides. Maltodextrin, starch, starch syrups, dextrins, proteins, gums and pectins are commonly used as carriers in spray drying. The content of a carrier in a liquid feed is usually at least 50% based on solid weight content [Johari et al. 1987, Hebbar et al. 2008, Jedlińska et al. 2019]. Hebbar et al. [2008] obtained honey powder with dextrin, maltose and anti-caking agents and high honey content of 52% that was characterised by good quality with its specific taste and aroma. Beside the low T_{σ} of honey sugars, its high viscosity is another issue that need to be dealt with for a convenient spray drying process. That viscosity is due to the relatively low water content of honey which can reach up to 20 wt% accompanied with high sugar content. That makes the process of spray drying of honey technically difficult. Therefore, dissolution (dilution) of honey with the appropriate diluent (commonly water) is indispensable. However, following todays' trends that are focused on whole foods with low glycemic index and that can also be labelled as "clean", maltodextrin with its high glycemic index, is becoming less and less desirable. These consumers' preferences force to search for alternative solutions [Stevenson et al. 2017].

Based on the above mentioned, the aim of this study was to examine the effect of the types of the diluent and carrier which are used for the dilution and delivery of honey on the process of spray drying of multiflower honey and the physicochemical properties of the obtained powders. Therefore, honey feed solution is prepared using maltodextrin and skimmed milk powder (separately) as carrier materials. In addition, liquid skimmed milk is going to be used as a novel diluent for honey before spray drying in order to study the effect of all these variants on the yield and physicochemical characteristics of the obtained honey powder.

MATERIAL AND METHODS

Materials

The raw material was multiflower honey from an apiary in Siemiatycze (Poland). The honey was collected from June to August 2018 and then stored at 17–18°C in closed glass jars. The raw material was liquefied and then stored in collective packaging in order to standardise the material.

Feed solutions preparation

Distilled water and Łaciate UHT milk 0.0% fat (SM Mlekpol, Grajewo, Poland) were used as diluents to prepare 30% (w/w) liquid feeds. Maltodextrin (MD) (DE 15-17) (Tate Lyle, Slovakia) and skimmed milk powder 1.25% fat (MP) (SM Mlekovita, Wysokie Mazowieckie, Poland) were used as carrier agents. The honey content in the feed solutions and the obtained powders ranged from 40 to 70% solids (Table 1).

- Table 1. Variants of spray dried honey with maltodextrin (MD) and skimmed milk powder (MP) used as carrier agents
- Tabela 1. Warianty suszonego rozpyłowo miodu z maltodekstryną (MD) oraz odtłuszczonym mlekiem w proszku (MP) jako nośnikami

	Roz	Diluent – water zpuszczalnik – woda	Dil Rozpuszcz	Diluent – skimmed milk Rozpuszczalnik – mleko odtłuszczone		
Carrier Nośnik	variant wariant	ratio of honey solids to carrier solids stosunek s.s. miodu do s.s. nośnika	variant wariant	ratio of honey solids to carrier solids stosunek s.s. miodu do s.s. nośnika		
	40MDW	40 : 60	40MDM	40 : 60		
MD	50MDW	50 : 50	50MDM	50 : 50		
	60MDW	60 : 40	60MDM	60 : 40		
	50MPW	40 : 60	50MPM	40 : 60		
MP	60MPW	50 : 50	60MPM	50 : 50		
	70MPW	60 : 40	70MPM	60 : 40		

Spray drying

Honey solutions were spray dried in duplicate of 500 g samples from each variant on laboratory spray dryer MOBILE MINOR (GEA, Denmark) with rotating disc rotation's speed at 26,000 rpm. Feed solutions were pumped at feed ratio speed 0.20 mL·s⁻¹ and inlet/outlet temperatures were respectively 180°C and 80°C. Drying yield [%] of each variant was calculated as the ratio of solids content in obtained powder to the solids in the feed solution.

Viscosity

Viscosity of solutions was determined using a MARS40 rheometer (Haake, Thermo Scientific, Japan) with coaxial cylinder geometry. Samples were analysed at 25°C with operating shear rate from 0 to 100 s⁻¹. Reograms were analysed using empirical models, and the apparent viscosity was calculated as the relationship between shear stress and rate.

Particles size distribution and morphology

The particle size distribution was determined by laser light diffraction using a 1190 instrument (CILAS, France). The powder was suspended in ethanol with at obscuration of 10%. The particle size was expressed as the median value of $D_{[43]}$.

The description of the powder particles morphology was prepared by an analysis of pictures taken with a scanning electron microscope (TM-3000 HITACHI) at 1,000× magnification.

Water content and activity

Water content was determined using oven method – approximately 1 g of powder was dried at 105°C for 4 h. Water activity (a_w) was measured using HygroLab C1 (Rotronic, Switzerland) at 24 ±2°C.

Bulk density and cohesiveness

Powders loose bulk density (*DL*) was determined by measuring the volume occupied by 10 g of powder. Tapped bulk density (*DT*) was measured using an automatic tapper STAV 2003 (Engelsmann AG, Germany) by determining the volume occupied by 10 g of powder after 100 taps. Hausner ratio (HR = DT / DL) was used to evaluate the cohesiveness and flowability of obtained powders.

Apparent density and loose bulk porosity

Apparent density (*Dapp*) was measured using helium pycnometer Stereopycnometer (Quantachrome Instruments, USA). Loose bulk porosity was calculated as: $\varepsilon L = 1 - (DL / Dapp)$.

Statistical analysis

All determinations were performed in three replicates for each sample. The results were statistically prepared using Statistica 12.5 (StatSoft Polska, Warsaw, Poland). A one-way analysis of variance ANOVA and a division into homogeneous groups using Tukey's test were performed at a significance level of $\alpha = 0.05$.

RESULTS AND DISCUSSION

Viscosity

The viscosity of the honey solutions ranged from 4.3 ± 0.1 to 11.0 ± 0.1 mPa·s (Table 2) and a significant effect of the composition of the solutions on the viscosity was found. The variants where milk powder and liquid skimmed milk which were used as carrier and diluent respectively, showed higher viscosity than the variants with the same ratio of honey to maltodextrin and water as a diluent.

Carrier Nośnik	Diluent – water Rozpuszczalnik – woda			Diluent – skimmed milk Rozpuszczalnik – mleko odtłuszczone		
	variant wariant	viscosity lepkość [mPa·s]	drying yield wydajność [%]	variant wariant	viscosity lepkość [mPa·s]	drying yield wydajność [%]
MD	40MDW*	5.3 ± 0.0^{cd}	$82.0\pm\!\!0.0^{bcd}$	40MDM*	$8.9\pm\!0.0^{g}$	99.7 ± 15.6^{d}
	50MDW*	$4.7 \pm 0.0^{\rm b}$	$62.0\pm\!\!5.6^{ab}$	50MDM*	$7.8 \pm 0.2^{\rm f}$	97.0 ± 2.5^d
	60MDW*	-	_	60MDM*	6.7 ±0.1 ^e	$93.2\pm\!\!2.2^{cd}$
MP	50MPW*	5.3 ± 0.0^d	81.1 ± 5.7^{bcd}	50MPM*	$11.0\pm\!\!0.1^h$	$95.2~{\pm}4.0^{d}$
	60MPW*	$4.9\pm\!\!0.3^{bc}$	70.1 ± 5.1^{ab}	60MPM*	$7.6\pm\!\!0.3^{\rm f}$	$93.3 \pm \! 0.4^{cd}$
	70MPW*	4.3 ±0.1 ^a	48.1 ± 5.2^{a}	70MPM*	6.7 ± 0.4^{e}	$67.2~{\pm}9.6^{ab}$

Table 2. Viscosity of honey liquid feeds before drying and drying yields of spray dried honey

Tabela 2. Lepkość roztworów miodu podawanych do suszenia rozpyłowego oraz wydajność procesu suszenia

*Shortcuts explained in the Table 1.

It can be observed that in both cases of used diluents, the viscosity decreased with the increase of the honey content in the solution. Samborska et al. [2015a] noted similar relationship when adding honey to the solution of gum arabic, which significantly reduced its viscosity.

Drying yield

As presented in Table 2, the powder 60MDW (60% honey content with maltodextrin as a carrier, water as a diluent) was impossible to produce due to its high honey content and low maltodextrin content. However, when liquid skimmed milk was used as diluent, it enabled to spray dry the variant containing 60% honey solids, where MD was used a carrier. In the other variants, the drying yield varied from 48.1 ± 5.2 to $99.7 \pm 15.6\%$ and it differed significantly depending on the type of the diluent and carrier. Moreover, when MP was used as a carrier, it enabled to increase the content of honey to 70% solids. It was not possible to obtain powders with such high honey content when MD was used as a carrier.

According to Bhandari et al. [1997], the spray drying can be considered successful when the drying yield is higher than 50%. It is worth noticing that only 70MPW's (70% honey content with skimmed milk powder as a carrier, water as a diluent) drying yield was below 50% (48.1 \pm 5.2%). The reason for this could be a large proportion of honey in the solution to milk powder (70:30), which significantly lowered the glass transition temperature (T_{o}) reducing the drying yield. The obtained results indicate that variants where MP was used a carrier, had higher drying yields sthan analogous variants where MD was used a carrier. Despite the lower T_g of lactose (101°C) than maltodextrin (188°C), better carrier properties of milk are associated with the presence of proteins in its composition.

The surface-active properties of proteins cause the formation of a membrane on the surface of sprayed droplets, which reduces the viscosity of the particles, while increasing the T_g of the external layer [Hebbar et al. 2008, Domian 2011, Samborska et al. 2015a].

Particles size and morphology

Scanning electron microscope (SEM) photos of the obtained powders presented in the figure, show that variants with 50% or less honey content in their composition, were characterised by a spherical structure. Particles of powders with MD and milk used as a diluent were more spherical and smoother compared to variants where water was a diluent. Moreover, conglomerated particles were not visible, unlike samples with water. This could be due to faster surface solidification caused by water evaporation from the layer of milk proteins that coated the surface of the droplets. The presence of milk in these variants indicates that the drying process is easier. The 70MPW and 70MPM variants consisted of conglomerated particles of irregular shapes and smooth surfaces. However, Samborska et al. [2019], after spray drying with the use of dehumidified air at low temperature (inlet/outlet 75/50°C) obtained more spherical and less conglomerated particles when milk was used as a diluent in 80% honey solutions with the addition of maltodextrin and nutriose as carriers.

The particles of the powders had a $D_{[3,4]}$ values from 19.3 ±3.9 to 70.3 ±4.3 µm (Table 3). Samborska et al. [2017] however, obtained less diverse results when producing honey powders with 30% honey content and MD (8–30 µm). In the variants with MD and



- Fig. SEM photos of obtained honey powders
- Rys. Zdjęcia SEM otrzymanych proszków miodowych

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Tabela 3. Właściwości fizyczne otrzymanych proszków miodowych: wielkość cząstek (D_[3,4]), gęstość nasypowa luźna (DL), gęstość utrzęsiona (DT), współczyn-nik Hausnera (HR), gęstość rzeczywista (Dapp), porowatość (EL)

	[%]	58.3 ±1.2ª	63.8 ±1.5 ^b	73.1 ±2.0 ^{ef}	66.6 ±1.6 ^{bc}	74.4 ±1.5 ^f	74.1 ±1.4 ^f
i milk czone mleko	$Dapp [{ m gcm}^{-3}]$	1.48 ±0.01°	$\begin{array}{c} 1.50 \\ \pm 0.00^{\mathrm{f}} \end{array}$	$\begin{array}{c} 1.51 \\ \pm 0.01^{g} \end{array}$	1.44 ± 0.00^{a}	1.45 ±0.00b°	$1.45 \pm 0.00^{\circ}$
	HR	1.22 ± 0.05^{a}	$\frac{1.31}{\pm 0.04^{ab}}$	1.46 ±0.06 ^{cde}	1.39 ±0.07 ^{bc}	$\begin{array}{c} 1.53 \\ \pm 0.05^{d} \end{array}$	1.51 ±0.09 ^{de}
nt – skimme nik – odtłusz	DT [g·cm ⁻³]	$\begin{array}{c} 0.75 \\ \pm 0.03^{\rm f} \end{array}$	0.71 ±0.01 ^{ef}	0.59 ±0.02 ^{bc}	0.67 ±0.04 ^{de}	0.57 $\pm 0.02^{ m abc}$	0.57 $\pm 0.02^{abc}$
Dilue: Rozpuszczał	DL [g·cm ⁻³]	$\begin{array}{c} 0.62 \\ \pm 0.02^{\mathrm{f}} \end{array}$	$0.54 \pm 0.02^{\rm e}$	0.40 ±0.03 ^{abc}	0.48 ± 0.02^{d}	$\begin{array}{c} 0.37 \\ \pm 0.02^a \end{array}$	0.38 ± 0.02^{a}
	$D_{[3,4]}$ [μ m]	70.3 ±4.3ª	57.3 ±4.8ª	19.3 ±3.9ª	24.5 ±7.7ª	30.0 $\pm 3.1^{a}$	40.1 ± 3.4^{a}
	variant wariant	40MDM*	50MDM*	60MDM*	50MPM*	60MPM*	70MPM*
	[%] 73	82.0 $\pm 0.0^{bcd}$	62.0 ± 5.6^{ab}	0.0	81.1 ±5.7 ^{bcd}	70.1 ± 5.1^{ab}	48.1 ± 5.2^{a}
	$Dapp [m g.cm^{-3}]$	1.52 ± 0.00^{g}	$\begin{array}{c} 1.54 \\ \pm 0.00^{\rm h} \end{array}$	I	1.44 $\pm 0.01^{ab}$	1.47 $\pm 0.00^{\rm d}$	$\begin{array}{c} 1.49 \\ \pm 0.01^{\mathrm{f}} \end{array}$
Diluent – water Rozpuszczalnik – woda	HR	$\begin{array}{c} 1.40 \\ \pm 0.04^{bcde} \end{array}$	$\begin{array}{c} 1.53 \\ \pm 0.10^{d} \end{array}$	I	$\begin{array}{c} 1.39 \\ \pm 0.04^{\mathrm{bc}} \end{array}$	1.40 ±0.04 ^{bce}	$\begin{array}{c} 1.39 \\ \pm 0.04^{\mathrm{bc}} \end{array}$
	DT [g·cm ⁻³]	$\begin{array}{c} 0.62 \\ \pm 0.01^{cd} \end{array}$	$\begin{array}{c} 0.76 \\ \pm 0.04^{\rm f} \end{array}$	I	0.60 ±0.02°	$\begin{array}{c} 0.54 \\ \pm 0.02^{ab} \end{array}$	$\begin{array}{c} 0.53 \\ \pm 0.01^{\mathrm{a}} \end{array}$
	DL [g·cm ⁻³]	0.44 $\pm 0.01^{cd}$	$\begin{array}{c} 0.50 \\ \pm 0.04^{de} \end{array}$	I	0.43 ±0.02 ^{bc}	$\begin{array}{c} 0.38 \\ \pm 0.02^{abc} \end{array}$	$\begin{array}{c} 0.38 \\ \pm 0.01^{ab} \end{array}$
	$D_{[3,4]}$ [μ m]	19.6 ± 1.1^{a}	36.5 ± 6.2^{a}	I	24.0 ± 1.4^{a}	$\begin{array}{c} 26.6 \\ \pm 1.7^a \end{array}$	37.0 ± 3.1^{a}
	variant wariant	40MDW*	50MDW*	60MDW*	50MPW*	60MPW*	70MPW*
Carrier Nośnik			MD			MP	

*Shortcuts explained in the Table 1.

milk as a diluent (40MDM, 50MDM, 60MDM), it can be observed that with the increase of honey content, the size of the particles decreased. For other powders the relationship was opposite – with the increase in honey content, the particles size increased. This may be caused by particles conglomeration due to too high drying temperature compared to the materials' T_{g} , which in consequence created bridges between the particles and resulted in larger particles.

The particles size was not affected by the viscosity of the liquid feeds. It was found that both the honey content and the viscosity of the solutions influenced the particles size. Samborska et al. [2015b] concluded that the particles size could be as well affected by the chemical composition of the dried solutions. After adding the gum arabic to honey, they observed larger particles despite the decrease in solution viscosity.

Bulk density and cohesiveness

The loose and tapped bulk density (DL, DT) of honey powders ranged from 0.37 ± 0.02 to 0.60 ± 0.02 g·cm⁻³ and from 0.53 ± 0.01 to 0.76 ± 0.04 g·cm⁻³ respectively (Table 3). No significant influence of the type of diluent on the loose and tapped bulk densities was noticed. However, the influence of the type of the carrier was observed. Powders with MD as a carrier showed higher densities than powders in which the carrier was MP. Goula and Adamopoulos [2010] noticed as well that as the MD content decreased, the bulk density of orange juice powders increased from 0.15 to 0.30 g·cm⁻³. Maltodextrin had the effect of reducing the cohesiveness and particles' sticking.

Cohesiveness of obtained powders, determined as HR, ranged from 1.22 ± 0.05 to 1.53 ± 0.10 (Table 4). According to Hausner [1967], powders with HR value > 1.4 are characterised by high cohesiveness, whereas < 1.25 are defined as powders with good flowability. All the variants with MP as a carrier, as well as the 50MDW and 60MDM were classified as powders with cohesiveness. Moreover, with decreasing carrier content in the variants with MD, the powders were characterised by higher cohesiveness. It can be observed as well that the change of diluent to milk in the case of the 50MPM and 60MPM, caused a deterioration of the flowability of obtained powders.

Apparent density and loose bulk porosity

The obtained powders were characterised by apparent density from 1.44 ± 0.00 to $1.54 \pm 0.00 \text{ g} \cdot \text{cm}^{-3}$ (Table 3). In all variants an increase in apparent density with an increase in honey content was observed. Powders with MP as a carrier obtained lower apparent densities than variants where MD was used as a carrier. However, the effect of diluent on apparent density was not noted.

The loose bulk porosity ranged from 56.9 ± 0.5 to $74.4 \pm 1.5\%$ (Table 3). The results are similar with the values obtained by Sułek and Domian [2010] who stabilised emulsions with milk proteins with the addition of trehalose and maltodextrin (62–77%). It can be observed that with the decrease of the carrier content in the liquid feed, the loose porosity increased. Nevertheless, the effect of the diluent and the carrier on the porosity of powders was not noted.

Water content and activity

The water content (*W*) in obtained powders was in the range from 0.7 \pm 0.5 to 4.1 \pm 0.5%, the values differed statistically significantly (Table 4). Shi et al. [2013], who spray dried honey with the whey protein isolate and MD as carriers, obtained higher values (3.1–5.0%). It is worth emphasising that all powders were characterised by water content < 5% which ensures their stability. The use of MD resulted in almost six times lower water content of 50MDW powder than when using milk powder in the 50MPW variant. This could be caused by the ability of milk proteins to form coating layer on the surface of the powder particles, which lowered the effectiveness of water evaporation.

Carrier Nośnik	Diluent – water Rozpuszczalnik – woda			Diluent – skimmed milk Rozpuszczalnik – mleko odtłuszczone		
	variant wariant	W [%]	a_w	variant wariant	W [%]	<i>a</i> _w
	40MDW*	1.8 ± 0.4^{abc}	0.174 ± 0.003^d	40MDM*	1.1 ± 0.5^{ab}	0.103 ± 0.017^{ab}
MD	50MDW*	0.7 ± 0.5^{a}	0.164 ± 0.005^d	50MDM*	$2.1\pm\!\!1.2^{abc}$	$0.116 \ {\pm} 0.013^{b}$
	60MDW*	_	_	60MDM*	2.6 ± 0.8^{abc}	$0.154 \ {\pm} 0.030^{cd}$
	50MPW*	$4.1 \ {\pm} 0.5^{d}$	0.096 ± 0.007^{ab}	50MPM*	2.4 ± 0.7^{abc}	$0.097\pm\!\!0.017^{ab}$
MP	60MPW*	3.9 ± 0.5^{cd}	0.120 ± 0.004^{bc}	60MPM*	2.3 ± 0.7^{abc}	$0.079\ {\pm}0.005^{a}$
	70MPW*	3.7 ± 0.3^{bcd}	$0.099\pm\!0.014^{ab}$	70MPM*	2.5 ± 0.2^{abc}	0.116 ± 0.009^{b}

Table 4.	Water content (W) and water activity (a_w) of obtained honey powders
Tabela 4.	Zawartość wody (W) oraz aktywność wody (a_{w}) uzyskanego miodu w proszku

*Shortcuts explained in Table 1.

The water activity (a_w) of the powders ranged from 0.079 ±0.005 to 0.174 ±0.003 (Table 4). Similar results were obtained when producing honey powders with MD and whey protein isolate as carriers, apple juice powders with MD and honey powders with the addition of sodium caseinate as carrier [Domian and Bialik 2006, Shi et al. 2013, Samborska et al. 2015a]. Regardless of the used diluent, variants with MD had higher aw than variants with MP. It may be caused by the presence of the protein layer in MP powders.

CONCLUSIONS

Skimmed milk in its both forms, powder (carrier) and liquid (diluent), can increase the content of honey in the final sprayed product compared to the traditional ingredients like maltodextrin and water. Additionally, milk improves the product's yield, as well as the properties of the finished product. Honey powders with high honey content (over 60%) using skim milk powder as carrier, water and milk as diluents were characterised by acceptable physical properties. Therefore, it can be concluded that skimmed milk powder can be a satisfying alternative to maltodextrin in the process of honey spray drying.

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BADANIE ZASTOSOWANIA ODTŁUSZCZONEGO MLEKA JAKO NOŚNIKA I ROZPUSZCZALNIKA PODCZAS SUSZENIA ROZPYŁOWEGO ROZTWORU MIODU

Streszczenie. Maltodekstrynę i odtłuszczone mleko w proszku zastosowano jako nośniki, a wodę i odtłuszczone mleko jako rozpuszczalniki do przygotowania roztworu miodu wielokwiatowego poddawanego suszeniu rozpyłowemu. Zastosowanie mleka poprawiło wydajność produktu. Ponadto umożliwiło to zwiększenie zawartości miodu w otrzymanych proszkach do 70% s.s. Mleko jako nośnik, a także jako rozpuszczalnik miało pozytywny wpływ na zawartość wody, aktywność wody, gęstość, kohezyjność i morfologię cząstek proszku. Z uzyskanych wyników wywnioskowano, że mleko w proszku może być z powodzeniem stosowane jako nośnik w suszeniu rozpyłowym miodu zamiast maltodekstryny.

Słowa kluczowe: miód w proszku, miód wielokwiatowy, suszenie rozpyłowe, nośniki suszarnicze, mleko w proszku