

Spray Drying of Honey: The Effect of Drying Agents on Powder Properties

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The aim of this study was to investigate the possibility of honey spray drying with addition of maltodextrin and gum Arabic as drying agents. The influence of the concentration of the solution subjected to drying, the type and content of the drying agents upon the physical properties of obtained powders was examined. An attempt was undertaken to obtain powder with a honey content of more than 50% d.b. Spray drying of multifloral honey with the addition of maltodextrin and gum Arabic was carried out at inlet air temperature of 180°C, feed rate of 1 mL/s and rotational speed of a disc atomizer of 39,000 rpm. The properties of obtained powders were quantified in terms of moisture content, bulk density, Hausner ratio, apparent density, hygroscopicity and wettability. Using gum Arabic it was possible to obtain a product with a higher content of honey (67% solids) than in the case of maltodextrin (50% d.b.). However, the powders obtained with gum Arabic were characterised by worse physical properties: higher hygroscopicity and cohesion, and longer wetting time.

INTRODUCTION

Honey is consumed because of its unique taste and aroma as well as its numerous health-promoting properties. The treatment involving bee products constitutes a science-based branch of medicine referred to as Apitherapy [Alvarez-Suarez *et al.*, 2010]. However, honey in its natural form has several disadvantages as a result of high viscosity and density which cause difficulties in transportation and dosage [Cui *et al.*, 2008; Hebbar *et al.*, 2008]. It can also change its properties as a result of crystallization, which may contribute to the development of osmophile yeast and fermentation [Bhandari *et al.*, 1999; Hebbar *et al.*, 2008]. The use of powdered honey significantly reduces these problems. Honey powder, like powders formed after drying of fruit juices, may be intended for direct consumption, used as an additive to a range of food products such as yoghurts, drinks, sauces, edible coatings, as well as dietary supplements and therapy-supporting preparations [Rodriguez-Hernandez *et al.*, 2005; Gabas *et al.*, 2007; Shrestha *et al.*, 2007; Hebbar *et al.*, 2008]. Production of honey dry powder is, however, difficult mainly due to the high content of sugars and organic acids [Truong *et al.*, 2005; Rodriguez-Hernandez *et al.*, 2005; Zareifard *et al.*, 2012; Murugesan & Orsat, 2012]. These substances are characterised by low glass transition temperature T_g . At T_g the form of the components changes from the hard, brittle “glass” material of a very high viscosity (about 10^{12} Pa·s)

into the soft visco-elastic “rubbery” material with viscosity of about 10^6 – 10^8 Pa·s [Williams *et al.*, 1955; Bhanardi & Howes, 1999; Shrestha *et al.*, 2007]. The temperature at which this transformation occurs is characteristic for each substance and, for substances from the same group, increases with the molecular weight. In honey, particularly important is the high content of fructose and glucose because of low T_g values: 10°C (fructose) and 36°C (glucose) [Papadakis *et al.*, 2006; Sahu, 2008]. Juszczak & Fortuna [2006] report the T_g value of multifloral Polish honey at -50.7°C (moisture content 17.7 % d.b.). According to other researchers [Lazaridou *et al.*, 2004], this value is -37.2°C in the case of the Greek multifloral honey (moisture content 14.1 % d.b.), and between -33.6 and -51.1°C in the Indian nectar honey from different plant species (total solids from 76 to 88% d.b.) [Ahmed *et al.*, 2007].

The form in which the material occurs during spray drying results from the relationship between product temperature T_p and the temperature of glass transition T_g [Noel *et al.*, 1990]. The amorphous material has to appear in a glassy form in order to obtain a free-flowing powder. These relationships are shown in Figure 1 [Samborska *et al.*, 2011]. During spray drying of the material with a high content of sugars, material temperature in the drying chamber T_{pl} is usually higher than the glass transition temperature T_{gl} . As a result, the material exists in the visco-elastic rubbery state, namely in the form of syrup or sticky particles adhering to the walls of the chamber. Drying such a material into a powder form is not possible [Bhanardi & Howes, 1999; Adhikari *et al.*, 2003; Hebbar *et al.*, 2008]. To enable the drying of this kind of material it is necessary to modify the pa-

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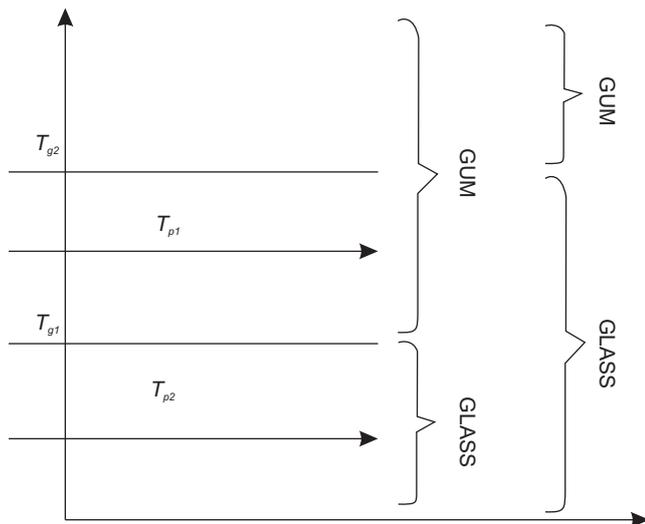


FIGURE 1. The influence of product temperature (T_p) and glass transition temperature (T_g) on the state of material during spray drying (descriptions in the text) [Samborska *et al.*, 2011].

rameters of drying or to prepare the material in such a way that $T_g > T_p$. At the glass transition temperature T_{g1} the material will occur in the glassy state during drying if its temperature does not exceed the T_{p2} . In practice this is difficult to achieve due to a low value of T_{g1} . The second option is to modify the composition of the material in a way to retain the temperature T_{p1} and achieve a higher value of glass transition temperature T_{g2} (where $T_{g2} > T_{p1}$). The way to increase the glass transition temperature of a material subjected to drying is to add substances such as starch, maltodextrin or gum Arabic [Truong *et al.*, 2004; Gabas *et al.*, 2007; Sahu, 2008; Jittanit *et al.*, 2010].

The available literature contains a few examples regarding the drying of honey and usually authors do not present the physical properties of honey powders, so in a current paper the results are compared and discussed also with the results regarding spray dried fruit juices. In most cases, the produced dried honey preparations contain up to 50% of honey in solid material. Cui *et al.* [2008] suggested a method of a microwave-vacuum drying of honey. The authors found that the best parameters for drying, resulting in the material with a water content of 3%, was the pressure of 30 mbar and film thickness below 8 mm. The content of sugars and aromatic substances remained unchanged after the drying process. Unfortunately, the authors did not specify the physical properties of the resultant product. Because of layer drying, it can be hypothesised that the final form of the material resembled hard solidified blocks rather than powder, which is not a beneficial form for further trade and use. Sahu [2008] vacuum dried liquid honey with three additives (maltodextrin, glycerol monostearate, tricalcium phosphate at different concentrations), spreading the mixes to a thickness of 3 mm. Vacuum (710–750 mm Hg) and temperature of 70°C were applied. Powder form was obtained by grinding in a hammer mill. Powders hygroscopicity (values between 4.32 and 12.3%) increased by decreasing the amount of maltodextrin and increased by increasing glycerol monostearate and tricalcium

phosphate. There was a decrease in degree of caking with an increase in the amount of all the three ingredients.

A beneficial form of dried honey may be obtained after spray drying with the addition of suitable drying agents [Hebbar *et al.*, 2008; Sahu, 2008; Samborska & Czelejewska, 2014; Samborska & Bieñkowska, 2013]. Spray drying is a method which, in addition to removing water (its primary objective), also allows the micro-encapsulation of substances vulnerable to external conditions, and there are many such biologically-active compounds present in honey. However, it is necessary to minimise the addition of drying agents so that the product is as close to natural honey as possible.

Yoshihide & Hideaki [1993] spray dried honey with the addition of antioxidants, carriers, partial dispersants, and dispersants, adjusting feed pH between 6.5–7.5 in order to reduce the thermoplasticity of sugary material during drying. Inlet and outlet air temperatures were between 120 and 200°C and between 70 and 120°C, respectively. The powder with the honey content of about 50% d.b. had good physical properties, pleasant flavour and taste. Samborska & Czelejewska [2014] used Arabic gum as a drying agent (honey solids: carrier solids, 1:1) and obtained powders of good physical properties: low water content and medium or good flowability. Samborska & Bieñkowska [2013] characterised the properties of spray dried honey preparations produced with the use of maltodextrin and dextrin as drying additives – those obtained with dextrin had higher hygroscopicity and worse solubility than those produced with maltodextrin. Takashi [1984] spray dried honey with the use of waxy starch as a carrier material at inlet temperature between 140 and 150°C (outlet 90–95°C). The resultant powder of final honey content less than 50% had a tendency to melt when exposed to air, hence the use of high barrier packaging materials was suggested. Hebbar *et al.* [2002] developed an improved method for the production of spray dried honey powder of honey content <52% solids, characteristic honey flavour, acceptable colour and of a free-flowing properties. Additives such as dextrin, maltose, and anticaking agent were used. Inlet and outlet temperatures (115–125°C and 80–85°C respectively) were lower than the conditions employed in the studies described above.

The aim of this study was to investigate the possibility of honey spray drying with the addition of maltodextrin and gum Arabic as processing agents. The impact of the concentration of the solution subjected to drying, the kind and content of the drying agent material upon the physical properties of resultant powders was examined. An attempt was also made to produce honey powder with a honey content of more than 50% d.b.

MATERIALS AND METHODS

Materials and preparation

Multifloral honey obtained directly from a local bee-keeper with a total solid mass concentration of $79.0 \pm 0.1\%$ d.b. (water content $19.6 \pm 0.1\%$ w/w) was used in the study. Maltodextrin 10 DE (MD, Peppes, Poland) and gum Arabic (GA, Hortimex, Poland) were used as drying agents. Experimental plan for spray drying of honey with maltodextrin, gum Arabic and the mixture of both drying agents is presented in Table 1.

Taking into account the solids content of honey and drying agents powders, 500 mL of solutions with the desired total solids concentration and the desired ratio of honey solids/drying agent solids were prepared. To obtain desired values it was necessary to dilute honey with water. The total concentration of solids in the solutions was 20 and 30% d.b.. The ratio of honey solids/MD solids was 1:2 and 1:1 (honey content 33 and 50% d.b.), while with the use of GA the ratio of honey solids/drying agent solids was 1:1 and 2:1 (honey content 50 and 66% d.b.). These values were selected in a preliminary study as the ratios at which it is possible to dry honey/drying agent/water solution, different for each additive substance. Additional experiments were also performed with the mixture of both drying agents MD+GA (1:1): total concentration of solids in the solutions was 20 and 30% d.b., ratio of honey solids/drying agent solids 1:1. Two runs were performed for each sample.

Viscosity analysis

Measurements were carried out on 16 mL samples using BROOKFIELD digital rheometer model DV-III (Brookfield Engineering Laboratories Inc., USA) in a range of shear stress corresponding to shear rates from 0 to 200 s⁻¹. The rheometer was equipped with a spindle type ULA that rotated in the sample-containing chamber at 25°C. All viscosity measurements, expressed in mPa×s, were performed in duplicate and averaged.

Spray drying

The drying was performed in a laboratory spray dryer Lab S1 (Anhydro, Denmark) with 1 m internal diameter, equipped with a rotating disc, using the following process parameters: inlet air temperature 180°C, raw material feed rate 1 mL/s, the rotational speed of a disc atomizer 39,000 rpm. During drying, the inlet and outlet air temperature was controlled and both values remained constant.

Powder analysis

Moisture content

Duplicate powder samples (approximately 1 g each) were dried at 105°C for 4 h [Chegini & Ghobadian, 2005]. The change in weight after treatment caused by water loss was expressed in percent by weight. The average of duplicate samples was calculated.

Bulk density

The powder loose (d_l) and tapped (d_t) bulk densities were measured using an automatic tapper STAV 2003 (Engelmann AG, Germany) by determining the volume occupied by 100 g of powder (tapped density after 100 taps).

Cohesiveness

Cohesiveness of the powders was evaluated based on Hausner ratio (HR), calculated from the loose (d_l) and tapped (d_t) densities of the powder: $HR = d_t/d_l$.

Apparent particle density

Apparent density of the spray dried powders was analysed using helium pycnometer Stereopycnometer (Quantachrome Instruments, USA). The pycnometric particle den-

sity was determined by measuring the volume occupied by a known mass of powder which is equivalent to the volume of displaced helium gas.

Hygroscopicity

Approximately 1 g samples of powder were placed in a desiccator under the following conditions: 25°C and 75% relative humidity (saturated NaCl solution) [Greenspan, 1977]. The gain in weight of the samples was measured during 2 h in 30 min intervals. Hygroscopicity has been expressed as the amount of water adsorbed by 100 grams of powder [Goula & Adamopoulos, 2010].

Wettability

Wettability was determined according to Jinapong *et al.* [2008] with some modifications. 100 mL of distilled water was poured into 800 mL beaker. A glass funnel was placed inside the beaker, with the height between the bottom of the funnel and the water surface of 100 mm. The lower opening of the funnel with the diameter of 40 mm was blocked by a test tube. The powder sample (10 g) was placed around the test tube and then the tube was lifted while the stop watch was started at the same time. The time necessary to wet all powder particles was recorded (visually assessed as when all the powder particles penetrated the surface of the water).

All analyses were performed in duplicate immediately after drying and the averages of these measurements were recorded.

Statistical methods

Analysis of variance was performed in order to determine whether the differences between obtained values were statistically significant. If the P-value of the F-test was less than 0.05, there was a statistically significant difference between the means at the 95.0% confidence level. To determine which means are significantly different from others the Multiple Range Tests were performed.

RESULTS AND DISCUSSION

Viscosity

Viscosity measurements showed that all the feed solutions followed a Newtonian behaviour, no shear-thickening or shear-thinning effects were observed under the applied measurement conditions (25°C, spindle type ULA, range of shear stress corresponding to shear rates from 0 to 200 s⁻¹). The estimated viscosity values ranged from 1.9 to 50.4 mPa×s, and were higher for GA mixtures than MD at the same concentration (Table 1). Solutions of lower solid concentration and higher honey content had lower viscosity. In most of the published papers, honeys are reported to be Newtonian fluids [Bhanardi *et al.*, 1999]. Popek [2002] presented the dynamic viscosity of aqueous 20% solutions of honeys belonging to the variety types: the values varied between 1.525 and 1.755 mPa×s. It is also generally recognized that dispersions of maltodextrin and gum Arabic are Newtonian in nature at concentrations of >10% [Meer, 1980; Dokic *et al.*, 1998; Mothe & Rao, 1999]. In the work presented by Shi *et al.* [2013], an increase in MD total solid ratio in the feed solution (water/MD/Capilano Natural Australian

honey solution) from 0 to 39.5% led to a negligible increase in viscosity from 6.00 to 8.00 mPa·s.

Production of honey powder

As described in the theoretical part, due to very low glass transition temperature of natural honey, the spray drying of honey without any additives is impossible. The first drying agent used in order to facilitate the drying of honey through increasing T_g was 10 DE maltodextrin (MD). Based on literature data the experiments began with drying of an aqueous solution of honey and MD in which the ratio of solids derived from honey to the solids originating from the drying agent was 1:2 and the overall dry matter concentration was 30 and 20 % d.b. These dryings proceeded smoothly and once they had been completed a significant amount of powder was obtained directly in the cyclone. It was also possible to recover the remaining powder from the walls of the drying chamber. Next, the MD addition in relation to honey solids was reduced so that the honey/drying agent solids ratio was 1:1. After these dryings, the quantity of powder in the cyclone was much smaller, the main bulk was recovered after cleaning the walls of the chamber. Also much greater sticking of chamber walls occurred and this level of MD was considered minimal to obtain powdered honey. Similar observations were presented by Papadakis *et al.* [2006] who spray dried raisin juice concentrate with the addition of MD 21 DE, 12 DE and 6 DE. Decreasing the content of MD in relation to the honey solids led to the reduction in the recovery of feed solids in the produced powder. In case of maltodextrin 12 DE the minimum content of the drying agent was 50% d.b., like in the current study. Shrestha *et al.* [2007] presented a relationship between T_g and product recovery after spray drying of orange juice with MD 6 DE. An increase in MD level from 50 to 60 parts resulted in a significant increase in product recovery (from 22 up to 78%), which was also accompanied by a huge jump in T_g from 66.4 to

86.4°C. The authors suggested that the maximum concentration of orange juice that can be dried in conjunction with MD (6 DE) is 40%.

In the following experiments the drying tests were carried out with solutions of honey with GA. Tonon *et al.* [2009] and Telis & Martinez-Navarrete [2009] have presented the T_g of spray dried açai juice and grapefruit juice powder obtained with MD and GA. Values obtained with the use of GA were higher than with MD. Based on these data, suggesting better GA properties as an anti-sticky agent, it was decided to start experiments with the GA from 1:1 level of drying agent content in relation to honey solids. Observations of the drying process, the quantities of powder in the cyclone and the material remaining on the walls of the chamber confirmed these assumptions, because it looked similar to drying with the MD at the honey/drying agent ratio of 1:2. Subsequently, the honey content was increased in relation to GA content to the honey/drying agent ratio of 2:1. As in the case of MD, it resulted in a reduction of the obtained powder quantity and an increase of the material remaining on the chamber walls. However, in all cases the amount of powder was sufficient to carry out further analysis of their physical properties. To sum up, using GA it was possible to obtain a honey preparation with a higher content of honey (67% d.b.) than using MD (50% d.b.).

Since the GA is more expensive than the MD, it was decided to carry out an experiment designed to replace part of the GA in a solution of MD, drying the samples containing honey, at MD and GA in the ratio of 2:1:1. Overall, the ratio of honey to the drying agent in these solutions was 1:1 and the overall concentration of dry substance was 30 and 20 g/100 g like before. The course of drying and the amount of powders obtained did not deviate from the samples dried using the GA in a 1:1 ratio to honey, which showed the feasibility of replacing parts of GA with MD without reducing the efficiency of the process.

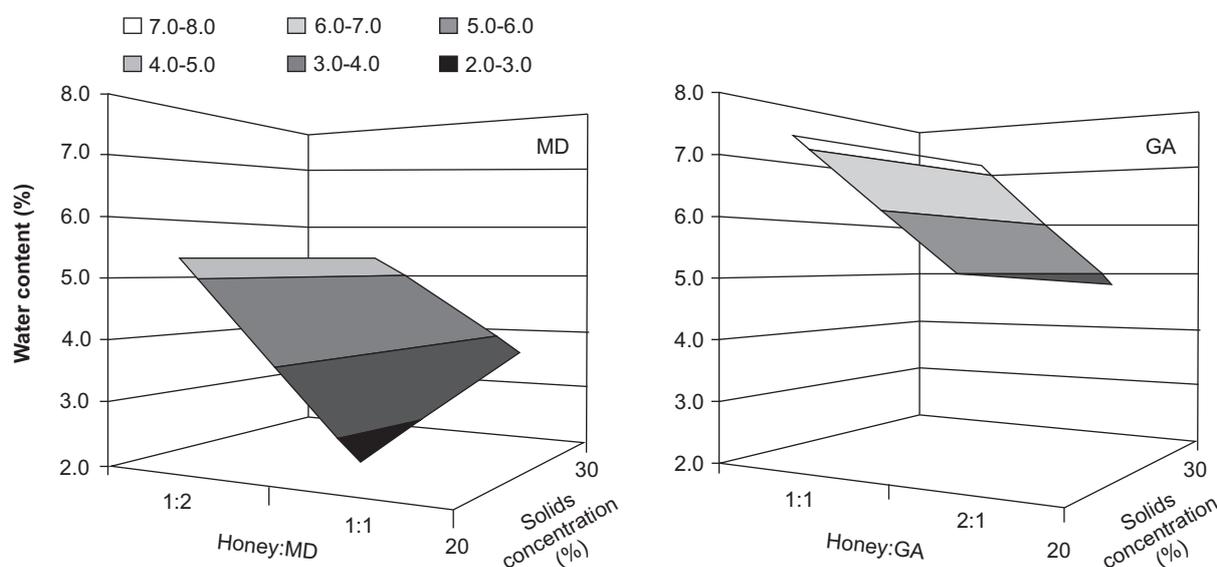


FIGURE 2. Surface plot showing the effect of honey content and solids concentration on water content of powders obtained after spray drying of honey solution with maltodextrin (MD) and gum Arabic (GA).

Physical properties of produced powders

Moisture content of powders

Water content of the obtained powders ranged from 2.7 to 8.6% (Figure 2). The differences between the obtained values were statistically significant (Table 2). The derived values were typical for powders obtained by spray drying of materials such as: multifloral honey (3.8 to 5.5% of water) [Ram, 2011], multifloral and rape honey spray dried with GA (7.1 and 7.3% of water) [Samborska & Czelejewska, 2014], juice from passion fruit with the addition of a mixture of MD (10 DE) and lactose (2.4 to 9.4% of water) [Ruiz-Cabrera *et al.*, 2009]. Vacuum dried honey powders obtained without the use of any additives had moisture contents values of 2.5% or less [Cui *et al.*, 2008].

Powders obtained using the MD usually contained less water than those containing GA. For example, the water content in experiment 4 with the use of MD was 2.7% and after the change of drying agent into GA (experiment 6) it increased to 7.2%. This relationship was also confirmed in the case of the pairs of experiments 3 and 5, which also had the same quantitative composition of the dried solutions but a different drying agent. The difference in the water content between these variants was statistically significant. A similar relationship was shown by Telis & Martinez-Navarrete [2009] who examined the properties of freeze-dried grapefruit juice. The same authors and Gabas *et al.* [2007] who studied the sorption properties of vacuum dried pineapple pulp found that the water content in monolayer was higher in case of dried material without additives, and after adding GA it was higher than in case of MD. Gabas *et al.* [2007] found that the presence of additives in pineapple pulp probably modified the balance of hydrophilic/hydrophobic sites, promoting a smaller amount of sorbed water. In the current work, this phenomenon was also observed, because the amount of water in powders obtained with MD was lower than in case of GA. This relationship can be connected with the viscosity of feed solutions, which affects the size of droplets produced during atomization, and thus the drying rate: the higher the liquid viscosity the larger the droplets and powder particles [Masters, 1991; Tonon *et al.*, 2008]. MD/honey solutions had usually lower viscosity than GA/honey solutions, so it can be assumed that the droplets were smaller, which could result in a higher drying rate leading to lower water contents in MD/honey powders. However, the correlation between the feed viscosity and water content was not always so clear, because the reduction of viscosity after decreasing of solid concentration from 30 to 20 g/100 g did not lead to the reduction of water content in the powders.

Powders obtained using the same drying agent with the same weight ratio of honey to the drying agent had a very similar water content regardless of the concentration of dry substance in the solution. Most of the results were laid in pairs, which is also clearly seen in Figure 2. This dependence has also been confirmed by the statistical analysis – in most cases there was no statistically significant difference between the water content of powders obtained from solutions of the same weight ratio of honey to the drying agent but with different concentrations of dry substance in a solution designed for drying.

The water content of the powders depended on the ratio of drying agent to the honey solids and thus on the content of the drying agent in a dried solution. The higher the content of the drying agent in the solution the higher the final water content of the obtained powders (compare *e.g.* experiments 1 with 3 and 5 with 7) what can be connected with increased viscosity of feed solutions at increased drying agent content. Similar results were achieved by Tonon *et al.* [2008] who studied the spray dried açai (*Euterpe oleracea* Mart.) juice and by Papadakis *et al.* [2006] who spray dried raisin juice concentrate with the addition of MD. Goula & Adamopoulos [2010] also found that the water content in the resulting powder increased along with an increasing MD content in the solution. This relationship was explained by the fact that for small water molecules it is difficult to diffuse through the larger maltodextrin molecules in the solution. Papadakis *et al.* [2006] spray dried a concentrated raisin juice at inlet air temperature of 110°C using the MD of varying dextrose equivalent (6 DE, 12 DE and 21 DE) and claimed that the maximum ratio of the content of the concentrated raisin juice to the added MD 6 DE was 67/33. Under these conditions, it was possible to obtain a stable powder with satisfactory physical properties.

Powders obtained after drying with mixed drying agents were characterised by water content at a level equal to or higher than recorded in powders with GA at the content of 1:1 in relation to the honey. The water content of powder derived from a solution of 30% d.b. was significantly higher than that of a 20% d.b. solution.

Powders loose and tapped bulk density

Loose bulk density of powders obtained with MD ranged from 0.33 to 0.55 g/cm³, with GA – from 0.31 to 0.47 g/cm³, and with MD+GA – from 0.47 to 0.58 g/cm³. Samborska & Bieñkowska [2013] after spray drying of multifloral honey with MD and dextrin reported loose bulk density of powders between 0.41 and 0.56 g/cm³, while in a study by Goula & Adamopoulos [2010] the bulk density of powders obtained after spray drying of orange juice with the addition of MD ranged from 0.14 to 0.41 g/cm³.

The type of the drying agent had a significant impact on the bulk density of powders. Comparison of the samples with the same quantitative composition but different drying agent type (*e.g.* in the following pairs of experiments: 3,5 and 4,6) reveals that a lower bulk density was observed in those samples which were dried using GA in comparison to those dried with MD. As shown in the Table 1, GA/honey mixtures had higher viscosity than MD/honey, which could lead to the production of larger droplets and particles, and this parameter is known to be negatively correlated with the bulk density: as typically, bulk density increases upon particle size decrease [Goula & Adamopoulos, 2005; Grabowski *et al.*, 2006].

In the case of powders dried using the same drying agent these results were correlated with water content, *i.e.* powders with lower water content had higher bulk density. Janiszewska *et al.* [2008] argued that the higher water content in powders causes their gathering into larger aggregations, which causes more empty voids between particles. This, in turn, results in a reduction of bulk density.

Differences in the loose density of powders with the same ratio of honey to the drying agent content but different initial concentrations of the solutions were not statistically significant.

The relationship of honey to the drying agent had the greatest impact on the value of loose bulk density – the lesser drying agent (in terms of weight) to honey ratio in the dried solution, the higher the bulk density of powders. This relationship can be seen by comparing samples 1, 2 with samples 3, 4 (MD) or samples 5, 6 with 7, 8 (GA). According to Goula & Adamopoulos [2010], the more sticky nature of a powder is associated with a high bulk density, thus the powders of smaller drying agent content, being more sticky, are characterised by a higher bulk density. In addition, increasing the amount of a drying agent may cause increasing amounts of air trapped between the particles, which is associated with skin-forming properties of MD. Kwapinska & Zbicinski [2005] found that particles of material with skin-forming properties often contain air bubbles from raw materials or adsorbed during spraying. Therefore, the greater the content of the drying agent with air bubbles the lower the bulk density of powders. Results reported by Shrestha *et al.* [2007] also confirm this relationship – the powder obtained by spray drying of orange juice was characterised by increasing bulk density along with decreasing amount of drying agent.

Goula & Adamopoulos [2010] observed that a loose bulk density is strongly linked to the water content of the powder. The higher the water content of the powder the more particles are combined into larger clusters resulting in voids occurring between them, which in turn results in a loose bulk density reduction. Results obtained in our study support this conclusion.

Bulk density of powders obtained after drying using a mixture of drying agents was in homogenous group b (20% d.b.) and group c (30% d.b.), *i.e.* it remained at medium and high level. These values were similar to those obtained after drying with individual drying agents with their lower content in relation to honey – 1:1 ratio in the case of MD and 2:1 ratio in the case of GA.

Cohesiveness

Hausner ratio (HR) values of the produced powders ranged from 1.05 to 1.29 (Table 2). According to the classification given by Geldart *et al.* [1984], powders with HR values below 1.25 are referred to as free flowing with low cohesiveness, while powders with HR values within the range of 1.25–1.4 are of average cohesiveness, and powders with HR higher than 1.4 are highly cohesive. Cohesiveness of powders determines their consistency and flow properties – the lower the cohesiveness the better the flowability of powders [Domian & Poszytek, 2005]. These results agree with previous studies on honey spray drying: Samborska *et al.* [2011] (multifloral honey with MD, HR value 1.26), and Samborska & Czelejewska [2014] (multifloral and rape honey with GA, HR values 1.29 and 1.24).

The Hausner ratio depended significantly on the type of drying agent content and the weight ratio of honey to the drying agent in the dried solution. By comparing 1:1 samples obtained from MD with those 1:1 from GA both with 30 and 20% d.b., it can be concluded that the powders with MD were characterised by a Hausner ratio lower than that

TABLE. 1. Experimental plan for spray drying of honey with maltodextrin, gum Arabic and the mixture of both drying agents.

Experiment	Drying agent: MD, GA	Honey/ drying agent ratio	Solids concentration (% d.b.)	Viscosity (mPa×s)
1	MD	1:2	30	8.7±0.2 ^{d1}
2	MD	1:2	20	2.2±0.3 ^{ab}
3	MD	1:1	30	4.5±0.4 ^{bc}
4	MD	1:1	20	1.9±0.4 ^a
5	GA	1:1	30	50.4±0.9 ^h
6	GA	1:1	20	13.2±0.5 ^c
7	GA	2:1	30	23.8±0.3 ^g
8	GA	2:1	20	5.0±0.3 ^c
9	MD+GA (1:1)	1:1	30	19.1±1.7 ^f
10	MD+GA (1:1)	1:1	20	4.1±0.1 ^{bc}

MD – maltodextrin, GA – gum Arabic

with GA, which indicates their better flowability. Reduction of the drying agent content in dried solutions affected the HR reduction of powders obtained from both MD and GA, *i.e.* powders with a higher content of honey had better flowability. Two powders of the worst flowability were those obtained with the use of GA – honey/ drying agent ratio 1:1 (experiments 5 and 6) – HR value higher than 1.25

The concentration of solids in dried solutions had no significant effect on the Hausner ratio because most samples dried using the same drying agent (at the same weight ratio) were in the same homogeneous group regardless of the concentration of solids in the initial solution.

On the basis of these results, it can be concluded that in terms of flowability the best powders were those obtained with the use of MD – honey/MD ratio 1:1 (experiments 3 and 4), followed by those obtained with the use of GA – honey/GA ratio 2:1 (experiments 7 and 8). Those were samples with a higher content of honey in dried solutions.

The comparison of Hausner ratio values of the obtained powders with their other physical properties reveals a clear relationship between HR and water content in the powders. The comparison of powders with the same drying agent showed clearly that the greater the water content in the powders the higher was the Hausner ratio. The Hausner ratio was also correlated with loose bulk density of the powders. Higher loose bulk density of powder corresponded with a lower Hausner ratio. This indicates that the powders with higher loose bulk density were the ones with lower cohesiveness and better flowability.

Powders obtained after drying using a mixture of drying agents in comparison to other powders were characterised by the average level of cohesiveness. However, the HR value below 1.25 indicated a low level of cohesiveness and good flowability of these powders.

Hygroscopicity

Hygroscopicity of the analysed powders was high and ranged from 65 to 145 g/100 g powder. Statistical analysis showed a division of values into three homogeneous groups. In the first group, with the lowest values of hygroscopicity,

TABLE 2. Outlet air temperature during drying and physical properties of dry product.

Experiment ¹	Outlet air temperature (°C)	Water content (%)	Loose bulk density d'_l (g/cm ³)	Tapped bulk density d'_t (g/cm ³)	Hausner Ratio HR	Apparent density (g/cm ³)	Hygroscopicity (g/100 g)	Wetting time (s)
1	66.0±0.5	5.4±0.2 ^c	0.33±0.01 ^a	0.41±0.03 ^a	1.24±0.03 ^c	1.53±0.01 ^{de}	85±4 ^a	54±3 ^b
2	67.0±0.7	5.4±0.1 ^c	0.33±0.01 ^a	0.38±0.02 ^a	1.18±0.01 ^b	1.45±0.01 ^b	72±3 ^a	57±2 ^b
3	69.0±0.5	3.7±0.4 ^b	0.55±0.01 ^c	0.57±0.02 ^c	1.06±0.02 ^a	1.48±0.01 ^{bc}	65±4 ^a	6±0 ^a
4	71.0±0.5	2.7±0.2 ^a	0.55±0.02 ^c	0.57±0.04 ^c	1.05±0.01 ^a	1.41±0.02 ^a	75±16 ^a	4±0 ^a
5	70.0±0.7	7.2±0.1 ^d	0.34±0.02 ^a	0.39±0.04 ^a	1.26±0.09 ^c	1.61±0.01 ^g	145±7 ^c	120±1 ^c
6	72.0±0.7	7.2±0.3 ^d	0.32±0.01 ^a	0.43±0.04 ^{ab}	1.29±0.04 ^c	1.58±0.02 ^{fg}	120±8 ^b	120±1 ^c
7	69.0±0.7	4.7±0.3 ^c	0.45±0.02 ^b	0.49±0.04 ^b	1.11±0.01 ^{ab}	1.57±0.01 ^f	115±23 ^b	9±0 ^a
8	69.0±0.7	5.0±0.1 ^c	0.47±0.03 ^b	0.54±0.04 ^b	1.15±0.06 ^b	1.55±0.01 ^{ef}	122±13 ^b	9±0 ^a
9	70.0±1.4	8.6±0.4 ^e	0.58±0.01 ^c	0.70±0.01 ^c	1.20±0.04 ^b	1.50±0.01 ^{cd}	130±7 ^{bc}	5±0 ^a
10	71.0±1.4	6.9±0.2 ^d	0.47±0.02 ^b	0.56±0.05 ^b	1.21±0.01 ^b	1.50±0.01 ^{bc}	132±14 ^{bc}	17±0 ^a

¹Experiments descriptions as presented in Table 1. ^{a-g}Means with the same superscript within same column are not significantly different (P<0.05).

were the powders obtained using MD. The powders produced with the addition of GA and GA + MD were enrolled into two consecutive groups characterised by significantly higher values of hygroscopicity.

Hygroscopicity values obtained are much higher than in other studies regarding fruit powders. For example, Rodriguez-Hernandes *et al.* [2005], who spray dried cactus pear juice with the addition of 18 or 23% MD obtained the hygroscopicity of powders varying from 36.3 to 48.9% d.b. Goula & Adamopoulos [2010] analysed the powder obtained after spray drying of orange juice with the addition of MD 12 DE in a ratio of 1:1 and 1:4 (orange juice: MD) and obtained the hygroscopicity of 4.2–5.3 g/100 g of solids depending on the drying temperature. Values in the current study regarding powders obtained with the use of MD (65–85 g/100 g powder) are higher than those presented above, which may indicate much higher hygroscopicity of other ingredients of the powders derived from honey.

Tonon *et al.* [2008] found that MD concentration was the variable which affected the hygroscopicity of açai powders produced by spray drying. The lowest hygroscopicity values were obtained when the highest MD concentrations were used and this correlated with the fact that MD is a material of low hygroscopicity. Rodriguez-Hernandes *et al.* [2005] observed the same relationship working with spray dried cactus pear juice. However, Papadakis *et al.* [2006] did not observe a clear pattern for the differences between the powders of spray dried raisin juice concentrate and different juice:MD ratios. In the current study, these differences were also statistically insignificant.

Gabas *et al.* [2007] have investigated the water sorption properties of pineapple powder obtained by vacuum drying with the addition of MD and GA. The monolayer water content was of particular interest, since it could be taken as an indicator of the volume of water that can be strongly adsorbed to specific sites at the food surface. Samples containing MD

resulted in the value of the monolayer moisture content X_m between 5.8 and 6.9% (dry basis), whereas samples with gum Arabic showed X_m between 6.7 and 7.9% (dry basis). Lower values of monolayer moisture content reported in the case of the powder containing MD indicate that this material was characterised by lower hygroscopicity than GA, which corresponds to the results shown in the present paper. Telis & Martinez-Navarrete [2009] studying the sorption properties of grapefruit juice powder also found that for samples with additives the X_m values were lower than for pure grapefruit juice, being also lower for samples with MD than with GA. According to Perez-Alonso *et al.* [2006], this phenomenon is attributed to a combination of factors such as the conformation and topology of molecule and the hydrophilic/hydrophobic sites adsorbed at the interface.

Hygroscopicity of powders obtained through drying with MD+GA was at the same level as for the powders obtained with GA.

Apparent (absolute) density

Apparent (absolute) density of the obtained powders ranged from 1.41 to 1.61 g/cm³ (Figure 3). Both the drying agent type and the ratio of honey to the drying agent in the dried solution had a significant effect on the apparent density of powders and, in the case of MD, also the initial concentration of the dried solution (Table 2). The value of apparent density shows the internal porosity of particles – the more “compact” a particle is the higher the apparent density. Janiszewska & Witrowa-Rajchert [2007] found that the higher the initial concentration of the dried solution the greater the values of apparent density of powders (lower internal porosity). Powders discussed in this paper show such a relationship but the differences were statistically significant only in the case of powders obtained with MD. For both drying agents, the apparent density was correlated with the viscosity of feed solutions.

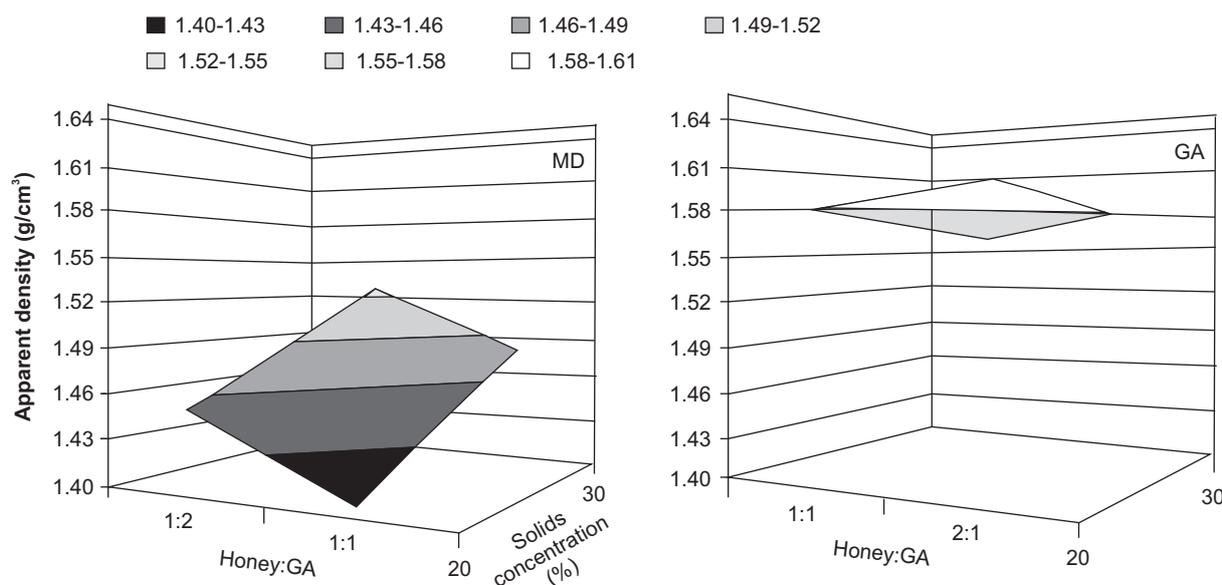


FIGURE 3. Surface plot showing the effect of honey content and solids concentration on apparent density (g/cm^3) of powders obtained after spray drying of honey solution with maltodextrin (MD) and gum Arabic (GA).

Abadio *et al.* [2004] obtained very similar values of apparent density of pineapple juice spray dried with MD (from 1.47 to 1.58 g/cm^3). However, they found that the increasing content of MD leads to a reduction in apparent density of powders, which has not been confirmed in the present study. Tonon *et al.* [2010] determined the absolute density of açai juice powdered by spray drying with addition of MD (10 DE and 20 DE), GA and tapioca starch, yielding values ranging from 1.49 to 1.53 g/cm^3 . In contrast to the current research, the samples produced with GA showed absolute density lower than these produced with MD 10 DE.

The powders dried with MD were characterised by significantly lower apparent density than those dried with GA. This conclusion can be reached by comparing samples 3 with 5 and 4 with 6. Janiszewska & Witrowa-Rajchert [2009] observed a similar relationship after microencapsulation of rosemary aroma by spray drying method. This indicates that the powders dried with MD were characterised by higher internal porosity.

Samples dried using MD + GA were characterised by an apparent density close to the apparent density of samples dried using MD. Powders with a higher content of honey had a lower apparent density.

Wettability

Wettability of the obtained powders ranged from 4.5 to 120 s (Table 2). Both the drying agent type and the honey content in the dried solution had a significant effect on this parameter. MD powders had better wettability than those with GA. In general, it can be also seen that wettability increased significantly (it means the wetting time was shorter) with an increase of honey content in the powder. It may be due to the higher sugar content in the powder, which causes that the moisture was adsorbed more rapidly [Jinapong *et al.*, 2008].

Wettability of the powders dried with mixed drying agents did not differ significantly from that recorded in powders with MD:honey content ratio of 1:1.

CONCLUSIONS

Spray drying of honey and production of powders using MD and GA as drying agents was conducted. It was found that the behaviour of GA as a drying agent was more beneficial because it allowed obtaining powders of higher honey content (67% of honey in powder solids) than MD (50% of honey in powder solids). However, it was found that the powders dried with GA were characterised by less favourable physical properties: higher water content, higher hygroscopicity and wetting time, higher cohesiveness indicating poorer flowability. Replacing half of the GA with MD helped to maintain high drying efficiency and thus obtaining powders with better flowability and wettability than those with GA. However, their hygroscopicity remained at equally high level. The aim of further research will be to produce honey powder with a higher content of honey and also more favourable physical properties.

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