# Influence of process conditions on the physicochemical properties of honey powder produced by spray drying

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**RESUMEN.** Se evaluó la influencia de las temperaturas del aire de entrada y salida sobre las propiedades físicoquímicas de la miel de abeja secada por atomización mediante un diseño factorial de tres niveles. Las variables independientes fueron temperatura del aire de entrada (130-170 ºC) y temperatura del aire de salida (75-85 ºC). El proceso se realizó en un secador por atomización rotatorio usando maltodextrina 9DE como soporte. Las variables contenido de humedad, cohesividad, higroscopicidad, solubilidad y contenido de hidroximetilfurfural fueron usadas como atributos de calidad de la miel en polvo. El contenido de humedad y el tiempo de solubilización disminuyeron con el aumento de las temperaturas del aire de entrada y salida, debido a que están directamente relacionados con la transferencia de calor y masas. La cohesividad, higroscopicidad y contenido de hidroximetilfurfural del polvo aumentaron con el incremento de ambas temperaturas. La optimización múltiple indicó que la temperatura del aire de entrada de 144 ºC y la temperatura del aire de salida de 85 ºC fueron las que contribuyeron al mínimo contenido de humedad (1,4 % másico base seca), cohesividad (1,34), tiempo de solubilización (62 s), higroscopicidad (25,4 g/100 g) y contenido de hidroximetilfurfural (15,3 mg/kg). Las partículas tuvieron un alto grado de uniformidad y una buena distribución de tamaño (diámetro promedio de partícula $D_{43} = 16.1 \pm 0.4$ μm).

**ABSTRACT.** The influence of inlet and outlet air temperatures on the physicochemical properties of spray dried honey was evaluated by using a three-level factor design. The independent variables were: inlet air temperature (130-170ºC) and outlet air temperature (75-85ºC). The process was carried out on a rotary atomizer spray dryer and with maltodextrin 9DE as carrier agent. Factors such as moisture content, cohesiveness, hygroscopicity, solubility and hydroxymethylfurfural content were used as quality attributes of the honey powders. Powder moisture and solubility time were negatively affected by inlet and outlet air temperatures, which are directly related to heat and mass transfer. Powders cohesiveness, hygroscopicity and hydroxymethylfurfural increased with increasing both temperatures. The multiple response optimisation indicated that an inlet air temperature of 144 ºC and an outlet air temperature of 85 ºC were predicted to provide the minimal moisture content (1.4 % wb dry basis), cohesiveness (1.34), solubility time (62 s), hygroscopicity (25.4 g/100 g), and hydroxymethylfurfural (15.3 mg/kg). The particles have a higher degree of uniformity regarding shape and good distribution (average particle size $D_{43} = 16.1 \pm 0.4$ μm).

## INTRODUCTION

Honey is an aromatic and sweet liquid derived from the nectars of plants, which is collected by honeybees and later modified and stored by them as a denser liquid, which is a complex mixture of sugars, enzymes, minerals, amino acids, and vitamins with associated health benefits. The sweetness, functional advantages and nutritional value of honey are encouraging food processors to be used it in different food products. However, honey, in its liquid and natural state, presents significant handling problems due to its viscosity and stickiness. There is a strong and constant consumer demand for dried honey that is convenient to be consumed or used in the food industry. Honey powders have many number of benefits over the liquid counterpart such as lower moisture, which allows its use directly into dry mixes, seasonings or dry coating of foods, reduced volume, reduced packaging, easier handling, transportation and longer shelf life.[1]

Dehydration by spray drying is used extensively in the food industry for a wide range of products in dry particulate form as powders.[2] However, the main problem during spray drying of sugar-rich foods such as honey is their thermoplastic behaviour. The chemical composition of honey is represented by components of low glass transition temperature (Tg), mainly fructose and glucose, and during drying they may either remain as syrup or stick on the dryer chamber wall.[3] This might lead to low product yields and operating problems, which can be minimized by the addition of high molecular weight additives to the product before atomizing (e.g. Arabic gum, starch, and maltodextrins) to prevent stickiness of product by increasing the Tg of the product during the process.[1]

There are some publications on the impact of drying methods,[1,4-6] types of carriers,[7,8] drying parameters[8] and drying aids[9-11] on the physicochemical properties of the honey powders. However, there is a lack of works investigating the influence of the drying temperatures on the properties of honey powder and the optimisation of these operational parameters by response surface methodology (RSM). RSM is an empirical modelling approach for determining the relationship between response variables with the various desired criteria and the significance of parameters affecting them. The main advantage of RSM is the reduced number of experimental trials needed to evaluate multiple parameters and their interactions. Nevertheless, this approach has not been applied to optimizing dryer operating parameters for honey. Therefore, the objective of this work was to study the influence of inlet air and outlet air temperatures by using RSM on the physicochemical properties during the microencapsulation of honey by spray drying, and to determine the optimum drying conditions for the production process.

## MATERIALS AND METHODS

### Materials

Multifloral honey (5 kg) derived directly from the apiary of the Apiculture Research Centre in Havana (2014 crop). It was characterized by a moisture content 20.0 ± 0.9 % wb, colour 104.0±4.0 mmPfund, conductivity 0.74 ± 0.02 m·S/cm, hydroxymethyfurfural content 7.19±0.09 mg/kg, free acidity 41.5 ± 0.9 meq/kg, reducing sugars 41.3 ± 0.8 g/kg, and diastase activity 37.62 ± 0.7 U Schade. Maltodextrin 9DE with moisture content of 6.0 ± 0.3 % was supplied by Glucidex (Lestem, France).

Spray drying of aqueous honey solutions with maltodextrin containing 30% m/m of dry matter (ratio of dry matter content coming from honey to the one originating from carrier, 1:1) was performed in a small-scale rotary atomizer Mobile Minor spray dryer (Niro Atomizer Ltd., Copenhagen, Denmark) under the following conditions: temperature of the inlet air temperature: 130, 170, and 180ºC; outlet air temperature: and 75, 80, and 85ºC; and atomising disk speed: 30,000 rpm. Ingredients (120 g honey + 102.8 g maltodextrin + 417.2 g distilled water) were mixed using a mechanical stirrer to obtain a uniform solution. The spray drying procedure was performed with 640 g of solution for each run. All formulations for spray drying were carried out in duplicates. The powders were collected from the cyclone separator, packed in polythene bags and stored in a desiccator at 20ºC before being submitted to triplicate analyses.

### Honey analyses

The liquid honey was analysed for moisture, total acid, conductivity, and diastase activity as described in Harmonised Methods of the European Commission of Honey.[12] Reducing sugars and hydroxymethylfurfural content were measured according to standard methods.[13] Colour was measured using a Hanna honey colour analyser HI-96785 (Hanna Instruments, Temse, Belgium).

### Powder analyses

Moisture content was determined in an electronic moisture balance Sartorious mod. MA35 (Goettingen, Germany). Relative standard error lower than 11%.

Loose ($d_L$) and tapped ($d_T$) bulk densities of powders were measured using a graduated cylinder by determining the volume occupied by 1 g of powder (tapped density after 50 taps).[14] Cohesiveness of the powders was evaluated in terms of Hausner ratio (HR), calculated from the bulk loose ($d_L$) and tapped ($d_T$) densities ratio: $HR = d_T/d_L$.

Hygroscopicity was determined by adapting a method previously proposed.[15] Powder samples (approximately 1 g) were placed in plastic vials, accurately weighed and placed in desiccators at 25 °C and equilibrated over saturated solution of sodium chloride with relative humidity of 75.3%. The samples were weighed after seven days equilibration process and the hygroscopicity was expressed as g of adsorbed moisture per 100 g of dry solids.

Solubility time of powders was measured by adding 2 g of the material to 50 mL of distilled water in a low form 100 mL-glass beaker. The mixture was agitated with a magnetic stirrer at 890 rpm (stirring bar 4 mm × 10 mm), the time required for the material to dissolve completely was recorded.[14]

Hydroxymethylfurfural analysis was performed according to the same procedures as in the case of fresh honey, but in order to obtain comparable results, the honey solution was prepared taking into account also the carrier content. Knowing the water content in the powder, and the fact that the dry powder solid was composed of 50% maltodextrin and 50% honey, the quantity of powder equivalent to 5 g of honey was calculated. The solution for further analysis was prepared with the use of this precalculated amount of powder. Hydroxymethylfurfural content was determined by the Winkler method with the use of *p*-toluidine and barbituric acid solutions, measuring resultant colour at 550 nm.[13]

Particle size and structure of powders were evaluated using a scanning electron microscopy 5130 SB (Tescam, Praga, Czech Republic). The samples were separately mounted on aluminum stubs using double scotch tape and sputter-coated with a thin layer of gold. Finally, each coated sample was transferred to the microscope where it was observed at an acceleration potential of 10 kV. Images at 2000x magnification were obtained with the software associated with the equipment, and the average particle size $D_{43}$ was calculated.

**Statistical analysis**

A three-level factorial design was used to design the tests for the spray drying of honey, considering two factors (independent variables): inlet air temperature and outlet air temperature, which dictated 12 experimental runs, including four central points. The final goal of RSM was to determine the optimum conditions for the system.[16] The analysis of variance, test for the lack of fit, determination of the regression coefficients and the generation of three dimensional graphs were carried out using the software package Design-Expert version 8 (Stat-Ease Inc., Minneapolis, MN).

## RESULTS AND DISCUSSION

**Response surface analysis**

Different physicochemical properties of the honey powders were evaluated which were produced at different inlet and outlet air temperatures. The average experimental results of these physicochemical properties are shown in Table 1. Table 2 shows the regression coefficients for the coded second order polynomial equation, the F value and the determination coefficients ($R^2$). Some non-significant terms were eliminated and the resulting equations were tested for adequacy and fitness by the ANOVA. The fitted models gave satisfactory adjustment of the reduced response models used for describing the response variables as function of inlet and outlet air temperatures.

Powders moisture content varied from 1.0 to 2.3% wb (Table 1), similarly to the range of the moisture contents of honey powder spray-dried with maltodextrin,[7,8,10] but were much lower than those spray drying honey with Arabic gum.[6] Moisture content was highly significant influenced by inlet and outlet air temperatures (Table 2). The coefficients of the first order terms with coded variables indicated that the moisture content decreased with the increase in inlet air temperature as well as outlet air temperature. This finding is typical for spray dried products, and it is related to heat and mass transfer.[17,18] On the other hand, other researchers reported no significant effect of an inlet air temperature on the moisture content of the spray-dried honey powder.[8] The inlet air temperature negatively affected powders moisture content. Higher outlet air temperatures are achieved with lower flow rates and this fact implies in a higher contact time between the feed and the drying air, making the heat transfer more efficient and resulting in greater water evaporation.

**Table 1.** Experimental data for response surface analysis of the effect of processing conditions on the quality of honey powder.

| Run | $T_I$ (ºC) | $T_O$ (ºC) | Moisture (% wb) | Loose bulk density (g/mL) | Tapped density (g/mL) | Cohesiveness | Hygroscopicity (g/100 g) | Solubility time (s) | HMF (mg/kg) |
|---|---|---|---|---|---|---|---|---|---|
| 1 | 150 | 80 | 1.5 | 0.4890 | 0.6846 | 1.40 | 23.00 | 59 | 16.30 |
| 2 | 150 | 85 | 1.2 | 0.4260 | 0.6006 | 1.41 | 27.30 | 50 | 15.60 |
| 3 | 150 | 80 | 1.4 | 0.5120 | 0.7065 | 1.38 | 23.30 | 64 | 15.81 |
| 4 | 150 | 75 | 1.9 | 0.4573 | 0.5944 | 1.30 | 22.59 | 70 | 15.00 |
| 5 | 150 | 80 | 1.5 | 0.4936 | 0.6663 | 1.35 | 23.10 | 61 | 15.31 |
| 6 | 170 | 80 | 1.1 | 0.4380 | 0.6526 | 1.49 | 27.80 | 53 | 18.26 |
| 7 | 130 | 80 | 2.1 | 0.5860 | 0.7325 | 1.25 | 21.60 | 92 | 14.85 |
| 8 | 150 | 80 | 1.5 | 0.4780 | 0.6501 | 1.36 | 23.80 | 62 | 16.00 |
| 9 | 170 | 75 | 1.8 | 0.5530 | 0.7907 | 1.43 | 22.63 | 52 | 17.66 |
| 10 | 170 | 85 | 1.0 | 0.4320 | 0.6307 | 1.46 | 32.10 | 49 | 18.30 |
| 11 | 130 | 85 | 2.0 | 0.5706 | 0.7018 | 1.23 | 21.80 | 90 | 14.62 |
| 12 | 130 | 75 | 2.3 | 0.5903 | 0.7083 | 1.20 | 20.90 | 93 | 13.17 |

$T_I$. inlet air temperature. $T_O$. outlet air temperature. HMF. hydroxymethylfurfural.

Loose bulk and tapped densities of powders had values typical for this kind of material after spray drying (Table 1), and the results are similar with those of honey spray dried with maltodextrin and whey protein isolate,[9] with maltodextrin and dextrin,[8] and with Arabic gum.[6]

Cohesiveness of powders determines their consistency and flow properties – the lower the cohesiveness the better the flowability of powders. Hausner ratio (HR) values of obtained powders ranged from 1.20 to 1.49 (Table 2). According to the classification given earlier,[19] powders of HR in the range from 1.2 to 1.4 are classified as intermediate cohesive. Only the powders dried with inlet air temperature of 170 ºC had HR>1.4 and then are classified as very cohesive powders and therefore with very poor flowability. These results are relatively higher than those of honey spray dried with maltodextrin and dextrin,[8] Arabic gum,[6] and Arabic gum and sodium caseinate;[11] but similar to those reported in the studies on spray drying of honey with maltodextrin.[10] The coefficients of the first order terms (Table 2) with coded variables indicated that the cohesiveness increased with the rise in inlet air temperature as well as outlet air temperature. Inlet air temperature was the variable that most affected powder cohesiveness. This finding could be explained by the changes in the loose bulk density, which is inversely related to HR, with inlet and outlet air temperatures. This dependence could arise from the fact that the loose bulk density is strongly linked to the moisture content of the powder. The higher the humidity, as such samples obtained after drying at lower inlet and outlet air temperatures, the more particles are combined into larger clusters, leaving open spaces between them, which in turn lowers the bulk density.[20]

**Table 2**. Coded regression coefficients for the physicochemical properties of honey powder.

| Coefficient | Moisture | Cohesiveness | Hygroscopicity | Solubility time | HMF |
|---|---|---|---|---|---|
| Intercept | 1.46 | 1.38 | 23.63 | 61.50 | 15.87 |
| $X_I$ | -0.42*** | 0.12*** | 3.04*** | -20.17*** | 1.93*** |
| $X_O$ | -0.30*** | 0.028* | 2.51*** | -4.33** | 0.45* |
| $X_I^2$ | 0.18** | -0.014 | 0.41 | 11.00*** | 0.67* |
| $X_O^2$ | 0.13* | -0.029 | 0.65 | -1.50 | -0.59* |
| $X_I X_O$ | -0.12* | 0.00 | 2.14*** | 0.00 | -0.20 |
| $R^2$ | 0.977 | 0.958 | 0.974 | 0.963 | 0.972 |
| F | 51.58*** | 27.24*** | 44.11*** | 31.33*** | 41.12*** |

$X_I$, $X_O$. coded inlet and outlet air temperature, respectively. HMF. hydroxymethylfurfural. *Significant at $p \leq 0.05$. ** Significant at $p \leq 0.01$. *** Significant at $p \leq 0.001$

Hygroscopicity of powders ranged from 20.90 to 32.10 g/100 g (Table 1). Honey powder is expected to be hygroscopic due to high content of sugars. A study of spray dried honey powder, with different ratios of maltodextrin and whey protein isolate,[9] at the same conditions of the present work, reported hygroscopicity range similar than those of the honey powders obtained in this work. Powder hygroscopicity was highly significant influenced by inlet and outlet air temperatures (Table 2). The lowest hygroscopicity values were obtained with decreasing both temperatures, which were the variables that also affected the powder moisture content, but in an opposite way. This indicates that the lower the powders moisture content, the higher their hygroscopicity, i.e., the greater their capacity to adsorb ambient moisture, which is related to the greater water concentration gradient between the product and the moist air. These results are in agreement with those published about spray drying of fruit pulp.[15,17,21]

The instant property of a powder is defined as its ability to dissolve in water. Hence, the ideal powder would dissolve quickly. The powders solubility time varied from 49 to 93 s (Table 1), similarly to the range of the moisture contents of honey powder spray-dried with maltodextrin and dextrin.[8] The coefficients of the first order terms (Table 2) with coded variables indicated that the solubility time decreased with the rise in inlet air temperature as well as outlet air temperature, being the first factor that most affected powders solubility time. This finding could be explained by the lower the moisture content of the powders the better is the solubility. Ambiguous results about the influence of the inlet air temperature were reported previously,[8] but the results of the present work are in agreement with those found in the spray drying of tomato pulp.[20]

Hydroxylmethylfurfural (HMF) content could be used as indication to see the degradation effect of heating during drying process on honey powder for each treatment. This furanic aldehyde is a derivative compound of fructose as a result of excessive heating of honey.[22] The HMF content of powders ranged from 13.17 to 18.30 mg/kg (Table 1), much lower than those of honey spray dried with maltodextrin[7] and with Arabic gum.[6] HMF content was highly significant influenced by inlet and outlet air temperatures (Table 2). Inlet air temperature was the variable that most affected HMF content. The coefficients of the first order terms with coded variables indicated that the HMF content increased with the rise in inlet air temperature as well as outlet air temperature, which is related to the greater destruction effect during the drying process. Nevertheless, any of the treatment values, even considering 1:1 the ratio of dry matter content coming from honey to the one originating from carrier, were below the maximum permissible statutory level of 40 mg/kg of honey.[23]

Numeric and graphic optimizations were carried out for the process parameters of` the honey powder. The desired goals for each response were chosen to be as follows: to minimize moisture content, cohesiveness, solubility time, hygroscopicity and hydroxymethylfurfural content. An inlet air temperature of 144ºC and an outlet air temperature of 85ºC were suggested to be optimal for spray drying of honey. Under such conditions, moisture content was estimated to be 1.4% wb, 1.34 cohesiveness, 62 s solubility time, 25.4 g/100 g hygroscopicity, and 15.3 mg/kg.

The optimization was checked according three runs made with the optimal parameters. The relative standard deviations among the experimental values and the calculated values by the models (results not shown) were lower than 10%.

Regarding the microstructure of the honey powder of the treatment obtained by spray dryer using the optimized parameters, it was noted that the particles have a higher degree of uniformity regarding shape with smooth and intact surfaces. The average particle size $D_{43}$ was 16.1 ± 0.4 μm, similarly to the range from 10-100 μm commonly found for this technology.[24] This result is lower than those of honey spray dried with maltodextrin and whey protein isolate,[9] but higher than those reported in the studies on spray drying of honey with Arabic gum.[6]

## CONCLUSIONS

Inlet and outlet air temperatures showed significant effect on all the studied physicochemical properties of spray dried honey. Increasing both temperatures led to lower powder moisture content and solubility time, which are directly related to heat and mass transfer. The increase of both temperatures also caused an increment in powder cohesiveness, hygroscopicity and hydroxymethylfurfural content. The recommended optimum conditions for spray drying honey, with addition of maltodextrin 9DE, were inlet and outlet air temperature of 144 °C and 85 °C, respectively. Morphological study of the honey powder obtained with these parameters revealed that the particles have a higher degree of uniformity regarding shape and an average particle size $D_{43}$ of 16.1 ± 0.4 μm. The results indicate that good quality honey powders with optimum physicochemical properties can be produced by spray drying, which demonstrates the great potential for the use of such powders in the food industry.

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