Spray drying of liquorice (*Glycyrrhiza glabra*) extract

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Revised: 21 August 2012 / Accepted: 3 September 2012 / Published online: 16 September 2012

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Abstract The objective of this work was to study the influence of spray drying conditions on the physicochemical properties of liquorice (*Glycyrrhiza glabra*) extract. The stickiness and hygroscopicity problems in the power were overcome by use of dextrose equivalent (DE12) and DE19 maltodextrins as drying agents. The inlet air temperatures of 110 °C, 120 °C, and 130 °C and maltodextrin concentrations of 10 %, 15 %, and 20 % (maltodextrin dry solids/100 g feed mixture dry solids) were the independent variables. Moisture content, bulk density, color change, hygroscopicity, acidity & pH, solubility were analyzed to determine the effects of spray drying conditions. Increases in inlet air temperature were caused an increase in yield, pH, solubility and a decrease in moisture content, bulk density, hygroscopicity, L*, a*, b*, acidity. Increases in maltodextrin concentrations were caused an increase in yield, L*, b*, acidity and a decrease in moisture content, bulk density, hygroscopicity, a*, pH, solubility. Increases in DE maltodextrins were caused an increase in bulk density, hygroscopicity, L*, pH and a decrease in yield, moisture content, a*, b*, acidity, solubility.

Keywords Liquorice · Liquorice powder · Maltodextrin · Hygroscopicity

Introduction

The liquorice plant has a long and storied history of use in both Eastern and Western cultures pre-dating the Babylonian and Egyptian empires. The genus name *Glycyrrhiza* is derived from the ancient Greek word for 'sweet root' (Gr. *glykos*

(sweet) + *rhiza* (root)), which was later Latinized to *liquiritia* and eventually to liquorice (Isbrucker and Burdock 2006). Commercially important sources are Spain, Iraq, Iran, Turkey, Russia and China, and although there are no known prohibitions against use of any species, variety or country of origin, some types are not sweet enough to have commercial value.

Liquorice root contains about 20 % of water-soluble extractives, and much of this typically 3–5 % of the root—is composed of glycyrrhizin. The sweet taste of the root comes from the substance glycyrrhizin, reputed to be 50 times sweeter than refined sugar.

Liquorice extract is produced by shredding and extracting the root with water in a steam extraction plant. The extracted liquor is filtered and then concentrated to produce a solid liquorice block or spray-dried to obtain liquorice powder.

Liquorice extract contains reducing and nonreducing sugars, starch, plant gums, resins, essential oils, inorganic salts and low levels of nitrogenous constituents such as proteins, individual amino acids, and nucleic acids (Isbrucker and Burdock 2006). Liquorice is largely used as a flavouring and sweetening agent, but has been proposed also for various clinical applications (Fiore et al. 2005).

Liquorice extract is added to certain types of chewing gum to insure a flexible texture and to certain chocolate candies to stabilize the fat dispersion. Especially in the United States much liquorice is used by the tobacco industry in cigarettes, cigars, smoking mixtures, chewing tobacco, and even snuff. Liquorice added to tobacco imparts a sweet taste and characteristic flavor and also enhances the mildness of a mixture. Liquorice in beer increases the foaminess of the beverage. In other drinks, such as root beer, porter, and stout, liquorice is added for flavor. Liquorice may also be used as a Brown coloring matter. Liquorice has been used medicinally for many centuries mainly to mask the bitter or acrid taste of other drugs or as a soothing remedy for affections of the respiratory tract. Liquorice is also a popular ingredient in herbal tea and in some botanicals, consumed to

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improve overall health and to alleviate a wide range of diseases (Ariño et al. 2007).

Spray drying is widely used for commercial production of dried fruits and vegetables. It is a well-established method for transforming a wide range of liquid food products into powder form. Spray-dried powders are suitable for transport and storage. The process results in powders with good quality, low water activity and easier transport and storage. The physicochemical properties of powders produced by spray drying depend on some process variables, such as the characteristics of the liquid feed (viscosity, particles size, flow rate) and of the drying air (temperature), as well as the type of atomizer. Therefore, it is important to optimize the drying process, in order to obtain products with better sensory and nutritional characteristics and better process yield (Tonon et al. 2008). Fruit juice powders obtained by spray drying may have problems in their properties, such as stickiness, hygroscopicity and solubility, due to the presence of low molecular weight sugars and acids, which have low glass transition temperature. Thus they can stick on the dryer chamber wall during drying, leading to low product yield and operational problems (Tonon et al. 2008). Stickiness is a physicochemical property of amorphous food powders. It depends on temperature and moisture content. If amorphous powders are at temperatures and/or moisture contents higher than what is called the powder sticky point and collide with each other. The sticky point temperature of amorphous powders has been related to the glass transition temperature, T_g , by many researchers (Spyridon et al 2006, Vardin and Yaşar 2012).

Various methods capable of producing a free-flowing fruit juice powder have been proposed: addition of drying aids (maltodextrins, glucose, soybean protein, sodium chloride, and skim milk powder), scrapping of dryer surfaces, cooling of the drying chamber walls, and admission of atmospheric air near the chamber bottom, allowing transport of the powder to a collector having a low humidity atmosphere (Goula and Adamopoulos 2010). High molecular weight drying aids have high glass transition temperatures. The lower DE of maltodextrin has the higher T_g value. The most frequently used additive is DE5 to DE30 maltodextrin (Spyridon et al 2006). Besides reducing powder hygroscopicity, such agents, normally used for microencapsulation, can protect sensitive food components against unfavorable ambient conditions, mask or preserve flavors and aromas, reduce the volatility and reactivity and provide additional attractiveness for the merchandising of food products (Tonon et al. 2008). Adhikari et al (2004) studied that the effects of addition of maltodextrin on the drying kinetics of simple sugars. In this study, lowering the drying rate occurred by addition of maltodextrin was reported. The reason of this was explained as the difficulties of water molecules to diffuse as compared to the larger maltodextrin molecules. Similar results were reported by Goula and Adamopoulos

(2008). The quality of spray dried powders depends on the operating parameters. Bhandari et al (1993) carried out a research to obtain powder from some concentrated juices. According to their study, the best results were obtained for a juice to MD ratio 65/35 for blackcurrant, 60/40 for apricot and 55/45 for raspberry respectively at inlet air temperatures between 90 and 160 °C. The effects of process variable are difficult to evaluate in general terms. The optimum process conditions can be determined by superimposing the contour plots. In the superimposed contour plots, the range of variables can be considered as the optimum range for best product quality. Vardin and Yaşar (2012) reported that 125–145 °C and 0.6–0.8 pomegranate juice/maltodextrin ratio was optimum process variable to produce acceptable dried products by using superimposing contour plots.

Drying models are classified into empirical and mechanistic models. The empirical models give some advantage of simple mathematical formulation and solution. However, these models cannot explain physics during drying and the application is limited since the models are only valid for particular drying conditions. On the other hand, the mechanistic models are derived based on phenomena during drying, which are however mathematically complex and sometimes too many parameters need to be determined before hand.

The aim of this study was to investigate the effect of different air inlet temperatures and addition of different DE maltodextrin, and addition of maltodextrin with different concentrations on the spray drying of liquorice extract. Also the powder properties (moisture content, bulk density, hygroscopicity, color, and acidity& pH, solubility) and spray drying performance (yield) was studied as parameters for optimization of process parameters.

Materials

The liquorice roots used were supplied from local markets (Gaziantep, TURKEY). Leaching at 25 °C by using the 1/3 as the ratio of liquorice/water was recorded as the optimum leaching procedure. The extract was then filtered through gauze and transferred to a bottle. Filtration was carried out to remove of fiber leafs at room temperature. Brix measurements were carried out by a digital refractometer (PTR 46X, Index Instruments). The extracts obtained were stored at refrigerator at 4 °C for one day. Filtered extracts was then concentrated in a rising film evaporator (Armfield FT 22, UK). The concentrate obtained was stored at refrigerator at 4 °C.

Methods

Maltodextrin was added accordingly to the liquorice extract. The mixture is then stirred till all maltodextrin was dissolved

and ready to spray dry. Co-current spray drying was performed using a laboratory scale spray drier (LabPlant SD-04, Huddersfield, England) with a standard 0.5 mm nozzle. The inlet air temperature varied from 110 °C to 130 °C and drying air flow rate was 0.35 m³/min. The feed flow rate and temperature were 5 mL/min and 40 °C respectively. The unit was self contained and supplied complete and ready for immediate operation. All major components were housed within a stainless steel cabinet. The temperature range (100–150 °C) was selected in a preliminary study to find optimum temperature without excessive powder stickiness on the chamber wall. Maltodextrin (%20, % 15, and % 10) was added according to the liquorice extract (20 °Bx). 19 DE and 12 DE maltodextrin were used. The dryer was washed with water at desired parameter settings for 10–15 min before spray drying process. Full factorial design was studied and spray drying experiments were carried out in one replicate. A total of 18 runs were conducted (Table 1). The powders produced were collected in a container, sealed, weighed and stored in dark.

Liquorise extract powder analysis

Spray dried powders were analysed for moisture content, bulk density, hygroscopicity, and acidity. Moreover, the powders produced with different maltodextrin concentration

Table 1 Experimental drying conditions for spray drying of liquorice extract

Experiment Number	DE*	Inlet Air Temperature (°C)	Maltodextrin Concentration (%)	Outlet Air Temperature (°C)
1	19	110	20	65–72
2	19	110	15	65–72
3	19	110	10	65–72
4	19	120	20	70–78
5	19	120	15	70–78
6	19	120	10	70–78
7	19	130	20	82–86
8	19	130	15	82–86
9	19	130	10	82–86
10	12	110	20	65–72
11	12	110	15	65–72
12	12	110	10	65–72
13	12	120	20	70–78
14	12	120	15	70–78
15	12	120	10	70–78
16	12	130	20	82–86
17	12	130	15	82–86
18	12	130	10	82–86

and at different inlet air temperature were analysed for solubility and color.

Yield was evaluated through determination of recovered product after spray drying, given by the ratio between the total recovered product mass and the mass of extract initially fed into the system. Process yield was calculated as the relationship between total solids content in the resulting powder and total solids content in the feed mixture (Tonon et al. 2008).

Each samples of liquorice powder (1 g each) were weighed and then dried in a drying oven at 80 °C for 3 h. The samples were taken out from the oven, cooled in a dessicator for 1 h and weighed. The drying and weighing processes were repeated until constant weights were obtained. The moisture content was expressed in terms of the percent wet basis (% w.b.) as 100×kg water/kg wet material (Rattes and Oliveira 2007). Duplicate samples were analysed and the mean reading was recorded.

The bulk density of the powder was measured by weighing out 1 g of the sample and placing it into a 10 mL graduated cylinder. This was tapped 10 times onto a cloth from a height of 10 cm. The volume was then recorded and used to calculate bulk density as g/mL (León-Martínez et al. 2010). The bulk density was calculated by dividing the mass of the powder by the volume occupied in the cylinder. Duplicate samples were analysed and the mean reading was recorded.

Samples (1 g) of each powder were placed in small glass covers, weighed and equilibrated over a saturated salt solution (PCl₃ solution; relative humidity of 90 %) in desiccators at 25 °C. After seven days, the samples were weighed and the hygroscopicity is expressed as g moisture/100 g solids (Kurozawa et al. 2009). Duplicate samples were analyzed and the mean reading was recorded.

The color of liquorice powder samples (L*, a*, and b* values) was measured with a HunterLab Colorimeter (Colorflex, USA) and results were expressed in accordance with the CIE Lab system (Ersus and Yurdagel 2007). Color attribute, hue angle (H*) was obtained from $\text{hue} = \arctg(b^*/a^*)$. Triplicate samples were analyzed and the mean reading was recorded.

The pH of reconstituted powder was measured by a pH meter. 10 mL of distilled water was added to 2 g of powder. The titratable acidity was determined as glycyrrhizic acid % (w/w) by titration with 0.1 N NaOH to a phenolphthalein end point at room temperature (Vardin and Yasar 2012). Duplicate samples were analyzed and the mean reading was recorded.

Fifty milliliters water was added to 2 g of powder. Then the mixture was stirred on the magnetic mixer. Solubility was determined as the time (second) that all of the powder was completely dissolved.

Experimental design and data analysis

A full factorial experimental design for two independent variables considered was inlet air temperature and maltodextrin concentrations. The levels of each variable were determined according to literature data and preliminary trials. The outline of experimental design with levels is presented in Table 1.

A polynomial model with second-order interactions (Eq. 1) was used to evaluate the main effects of the independent variables on each of the response variables:

$$z = a_0 + a_1x + a_2y + a_3xy + a_4x^2 + a_5y^2 \quad (1)$$

where z represents the response variables (yield, moisture content, bulk density, hygroscopicity, solubility, acidity, L^* and hue values), x is the inlet air temperature (110, 120 and 130 °C), y is maltodextrin concentration (20, 15 and 10 %), respectively, and a_0 , a_1 , and a_2 are the regression coefficients for the linear terms; a_3 is the interaction term; and a_4 and a_5 are the quadratic terms of the model. The multiple regression method (MLR) was used to adjust a polynomial model to the experimental data with a confidence level of 95 %. by non-linear regression analysis using the SigmaPlot 2000 (SPSS Inc., Chicago, IL, USA). The nonlinear regression analysis was carried with the Marquardt-Levenberg algorithm; this is a search method to minimize the sum of squares of the differences between the predicted and experimental data.

Moreover, spray drying parameters were optimized by using a convectional graphical method in order to obtain powder products with acceptable properties such as hygroscopicity, solubility, acidity, and bulk density. Predictive model (Table 4) was used to graphically represent responses as shown in Fig. 1a–b. The contour plots of all the responses were than superimposed and optimum region appeared. The contour plot (Fig. 3) was obtained as a result of overlaying of contour plots from which one can determine optimum process variables range.

Results and discussion

Mathematical modeling and statistical analysis

Regression coefficients and variance analysis of the second order polynomial model to evaluate the spray drying conditions on the physicochemical properties of powder shown in Table 4. The statistical analysis indicated that the proposed model fitted experimental data with R^2 at confidence level of 95 % ($p < 0.05$) and p values of each responses. For the yield, it was observed that, inlet air temperature is the most significant factor with respect to bulk density. This was

influenced by inlet air temperature, maltodextrin concentration and inlet air temperature-maltodextrin concentration interactions. Hygroscopicity was affected by the inlet air temperature. Inlet air temperature was also significant to derive the hue value.

Spray drying performance

It was first attempted to spray dry liquorice extract at 20 weight % solids with inlet air temperature from 100 °C to 150 °C. These attempts were unsuccessful due to powder stickiness problems. No powder was collected and toffee like deposits was formed on the main chamber and cyclone walls. Generally, low-DE maltodextrins have slower drying rates. This is attributed to the fact that the lower the DE of a maltodextrin, the higher the glass transition profile the more viscous the mixture. This leads to a lower evaporation rate due to the higher resistance to mass transfer. In addition, Rodriguez-Hernandez et al. (2005), who used maltodextrins as carrier agents during spray drying of cactus pear juice, reported that the lowest values of powder moisture content were obtained with the maltodextrin with the highest DE. According to them, the functional properties of maltodextrins depend on the polymerization degree, expressed as DE, and the lower the DE, the better binder agent the maltodextrin is. However, according to Goula and Adamopoulos (2008), the higher DEs caused the lower drying rates. This observation may be explained by the fact that high-DE maltodextrins develop stickiness slower and reach a state of non-adhesion slower than low-DE maltodextrins. Thus, we decided to try DE12 and DE19 maltodextrins as drying agents.

Minimum yield (~42 %) was found to be at inlet air temperature of 120 °C and maltodextrin concentration of 10 %. (Fig. 1). Above this point, inlet air temperature approaches the sticky temperature. Below this point, moisture content of powder increases.

The results of the yields are as shown in Table 2. Increasing temperatures led to higher process yield (Fig. 1a and b.), which can be attributed to the greater efficiency of heat and mass transfer processes occurring when higher inlet air temperatures are used. This is in agreement with the results published by Tonon et al. (2008), working with spray drying of acai (*Euterpe oleraceae* Mart.) powder. Inlet air temperature showed a positive effect on process yield, which can be attributed to the greater efficiency of heat and mass transfer processes and decrease probability of hitting the inadequate drying particles to the drying chamber wall when higher inlet air temperature are used. The similar results were shown by Fazaeli et al. (2012), Tonon et al. (2008), and Goula and Adamopoulos (2005). However Vardin and Yaşar (2012) reported that by increasing the inlet air temperature the process yield of pomegranate juice decreased. It is due to

Fig. 1 Effect of inlet air temperature and maltodextrin concentration on different physico-chemical quality characteristics of (a) Maltodextrin 12 DE and (b) Maltodextrin 19 DE

Fig. 1 (continued)

stickiness problems and it means that drying temperature is above their glass transition temperatures.

In this study the highest process yield (66 %) refers to powders produced with 20 % maltodextrin DE12 at inlet air temperature of 130 °C and the lowest drying yield (33 %) refers to powders produced with 10 % maltodextrin DE19 at inlet air temperature of 110 °C. The drying yield was increased with decreasing DE value of maltodextrin. Vardin and Yaşar (2012) and Fazaeli et al. (2012) reported similar results.

Powder properties

Moisture content

The powder moisture contents were analysed as between from 2.56 % to 5.63 % (wet basis). Figure 1a and b show the influence of inlet air temperature and maltodextrin concentration on powder moisture content. Increases in the inlet temperature resulted in decreases in the powder moisture content. This is because at higher inlet temperature, the heat transfer between particles is greater which will give a greater driving force for moisture evaporation. Thus, powders with less moisture content were formed. Osman and Endut (2009), Quek et al. (2007), Rattes and Oliveira (2007), Goula and Adamopoulos (2010), Ersus and Yurdagel (2007), and Tonon et al. (2008) also observed a reduction of powders moisture content with increasing air temperatures, studying the spray drying of roselle-pineapple juice, watermelon juice, sodium diclofenac, orange juice concentrate, black carrot and açai, respectively. An increase in

solids in the feed reduces amount of free water for evaporation. So that, a decrease in the moisture content of liquorice powder was obtained when the maltodextrin concentration increased. Similarly, Abadio et al. (2004) found that an increased concentration of maltodextrin reduced the moisture content of resultant pineapple juice powders. A similar result was also reported by Osman and Endut (2009) who carried out tests on roselle-pineapple juice powders. These findings could be explained by the fact that additional concentrations of maltodextrin resulted in an increase in feed solids and a reduction in total moisture for evaporation (Kha et al. 2010).

Higher maltodextrin dextrose equivalent caused an increase in powder moisture content (Fig. 1b). Goula and Adamopoulos (2010) mentioned that this may be due to the fact that high-DE maltodextrins develop stickiness slower and reach a state of non-adhesion slower than low-DE maltodextrins. The stickier a material is, the lower its drying rate.

Bulk density

The bulk density of liquorice powders varied from 0.18 to 0.323 g/mL (Table 2). Decreases in bulk density were observed with increase in inlet air temperature (Fig. 1a and b). Walton and Mumford (1999) observed similar trends for the bulk density. Kha et al. (2010); Goula and Adamopoulos (2010) also observed a reduction of powders bulk density with increasing air temperatures, studying the spray drying of Gac (*Momordica cochinchinensis*) fruit aril powder and orange

Table 2 Physico-chemical properties of liquorice extract powder

Run No	Yield (%) (d.b.)	Moisture Content (%) w.b.)	Bulk density (g/ mL)	Hygroscopicity (g/100 g) (d.b.)	Solubility (s)	pH	Acidity (%)
1	36	5.23±0.05	0.242±0.003	72±6	42±4	6.17±0.05	2.10±0.03
2	38	5.54±0.05	0.265±0.007	76±2	40±1	6.02±0.01	1.69±0.03
3	33	5.63±0.03	0.323±0.003	82±1	39±1	6.34±0.04	0.95±0.05
4	42	4.81±0.01	0.255±0.001	68±2	41±2	6.15±0.02	1.32±0.02
5	38	4.96±0.02	0.288±0.001	65±9	38±1	6.37±0.01	1.85±0.06
6	35	5.18±0.01	0.308±0.005	70±5	37±3	6.34±0.03	0.82±0.01
7	44	4.55±0.08	0.240±0.010	59±3	35±1	6.44±0.02	1.19±0.04
8	40	4.62±0.03	0.253±0.005	64±8	36±2	6.53±0.07	1.07±0.09
9	39	5.00±0.01	0.270±0.005	72±2	34±1	6.56±0.02	0.70±0.01
10	51	3.52±0.02	0.229±0.013	34±2	30±2	5.64±0.06	3.21±0.12
11	44	3.65±0.22	0.230±0.005	37±5	27±1	5.92±0.01	2.79±0.03
12	48	3.82±0.03	0.242±0.003	42±1	25±1	5.97±0.01	2.42±0.01
13	53	3.16±0.06	0.226±0.001	22±1	23±3	5.93±0.01	2.26±0.01
14	47	3.38±0.07	0.229±0.002	26±1	22±2	6.02±0.04	2.67±0.04
15	42	3.50±0.30	0.238±0.003	30±5	24±1	6.17±0.08	2.38±0.10
16	66	2.56±0.02	0.180±0.030	20±3	21±3	6.06±0.02	2.63±0.03
17	62	3.29±0.05	0.211±0.021	23±4	20±1	6.18±0.01	2.51±0.02
18	54	3.45±0.17	0.224±0.003	25±3	18±2	6.24±0.03	2.10±0.05

juice powders, respectively. Goula and Adamopoulos (2010) reported that evaporation rates are faster and products dry to a more porous or fragmented structure, there was a greater tendency for the particles to be hollow. Many researchers (Vardin and Yasar 2012, Tonon et al. 2008) have reported particle size in the range of 5–20 μm by the use of mini spray-dryers. These dryers employ pneumatic atomizer nozzles to produce droplets as small as possible with the purpose of achieving drying. On the other hand, particle agglomeration does occur in the product deposited on the dryer and cyclone walls. Bulk density is affected by the particle size. A higher proportion of small sizes increase bulk density. Increasing the drying air temperature produces a decrease in bulk density due to the increase in particle size. The higher drying rate obtained at higher drying temperatures that produce a higher ratio of surface to volume for the spray dried capsules, thus causing lower bulk density of the powders. This leads to the formation of vapor impermeable films on the droplet surface, followed by the formation of vapor bubbles and, consequently, droplet expansion (León-Martínez et al. 2010).

Bulk density showed a decrease with an increase in maltodextrin concentration (Fig. 1a and b). This effect may be attributed to the fact that maltodextrin addition minimizes thermoplastic particles from sticking. In addition, an increase in maltodextrin concentration may cause an increase in the volume of air trapped in the particles, as maltodextrin is a skin-forming material. Generally, an increase in the volume of trapped air causes a decrease in the apparent density of the particles and this apparent density primarily determines the powder bulk density (Goula and Adamopoulos 2010). Similar results were also reported by Vardin and Yasar (2012) who carried out tests on pomegranate juice powder.

An increase in maltodextrin dextrose equivalent resulted in an increase in bulk density (Fig. 1b). This can be attributed to the fact that the higher the maltodextrin DE, the lower its glass transition temperature and, as a consequence, the lower the elevation of the Tg of the liquorice extract–maltodextrin mixture is and the more stickier the mixture is (Goula and Adamopoulos 2010). Maximum bulk density (0.24 g/mL) was found to be at inlet air temperature of 120 °C and maltodextrin concentration of 10 % (Fig. 1). The reason of this maximum can be explained with sticky temperature and moisture content of powder.

Hygroscopicity

Liquorice powder is evidently hygroscopic. Spray dried particles can easily absorb moisture from the surrounding air and, unless necessary precautions are taken, the surface of the powder becomes sticky and powder caking occurs (Goula and Adamopoulos 2010). Moisture adsorption of the

spray dried powders at 25 °C and 90 % relative humidity after one week is shown in Table 2. Figure 1 shows the influence of maltodextrin concentration and inlet air temperature on hygroscopicity. Increases in inlet air temperature lead to higher powder Tg and, as a result, to lower hygroscopicity. Osman and Endut (2009) reported that the higher hygroscopicity of the powders produced at lower temperatures seems to be related to their higher moisture content. A major factor affecting powder stability is moisture content, since a small amount of water is able to depress the temperature enough to increase the mobility of the matrix during storage. Jaya and Das (2004) obtained similar results, working with the production of mango powder.

Increases in the maltodextrin concentration lead to higher powder Tg and, as a result, to lower hygroscopicity. According to Kurozawa et al. (2009), since the glass transition temperature increases with increase in molecular weight, the addition of maltodextrin to the feed solution contributed significantly to powder stability, increasing the Tg of the powder, and consequently reducing hygroscopicity and the stickiness. Maltodextrin is a material with low hygroscopicity, and confirms its efficiency as a carrier agent. This observation is similar to that reported by other researchers (Tonon et al. 2008; Goula and Adamopoulos 2010).

Higher maltodextrin dextrose equivalent caused an increase in hygroscopicity (Fig. 1b). Tg is related to chain stiffness and polymer chain structure; it increases as cross-link density increases. A lower Tg causes higher hygroscopicity in the spray dried powder, as lower molecular weight implies shorter chains and more hydrophilic groups. Goula and Adamopoulos (2010), León-Martínez et al. (2010) and Kurozawa et al. (2009), and also observed similar results, studying the spray drying of orange juice concentrate, nopal mucilage (*Opuntia ficus-indica*) and, chicken meat protein hydrolysate, respectively.

Solubility

The results of the solubility measurements for powders are shown in Table 2. A decrease in the solubility was observed with decreased inlet air temperature (°C), an increased DE maltodextrins, and increased maltodextrin concentration (Fig. 1). The higher temperature of the solvent is the faster its rate of dissolving and the greater its solubility. Abadio et al. (2004) also observed similar results. The moisture content influences powder properties such as solubility and bulk density. According to Fig. 2, solubility showed an increase with an increase in inlet air temperature. This is due to the effect of inlet air temperature on moisture content. However, bulk density and moisture content were higher and solubility was lower. The similar trends were reported by Tonon et al. 2008, and Vardin and Yaşar 2012.

Fig. 2 Effect of moisture content on solubility and bulk density of powder

Acidity and pH

The results of the acidity & pH measurements for powders are shown in Table 2. A decrease in the acidity was observed with an increased inlet air temperature ($^{\circ}\text{C}$), an increased DE maltodextrins, and decreased maltodextrin concentration (Fig. 1).

Color parameters

It is well known that high temperature can significantly decrease the quality of dried materials; e.g., by change of color. The results of the colour measurement for powders are shown in Table 3. L^* value measures the lightness of the sample, $+a^*$ measures the redness while $+b^*$ measures the yellowness. It was found that when inlet temperature increased, both the $+b^*$ values and the $+a^*$ values decreased (Table 2). The possible explanation for this phenomenon is that carrying out the spray-drying process caused rapid pigment oxidation, non-enzymatic browning. Therefore, the spray drying conditions at high temperature resulted in a high loss of red colour due to thermal degradation of pigments (Kha et al. 2010). Overall, the lightness of the powders reduced. This means that the colour of the powders has become darker at higher inlet temperature. This is related with the sugars liquorice contain. Sugars could contribute to browning of the powders at higher inlet temperature. (Quek et al. 2007) also observed similar results, studying the spray drying of watermelon juice and tomato products, respectively. Hue angle measures the main properties of the colour values. Overall, the lightness of the powders reduced and the hue angle of the powders decreased (Fig. 1).

Table 3 Color parameters of liquorice extract powder

Run No	Color			
	L^*	a^*	b^*	Hue*
1	59.4±0.09	3.0±0.07	28.7±0.03	84.1±0.13
2	59.5±0.09	2.8±0.03	27.8±0.02	84.2±0.07
3	55.4±0.11	3.5±0.02	27.9±0.04	82.9±0.04
4	59.1±0.26	3.2±0.09	28.6±0.07	83.7±0.17
5	57.7±0.48	3.2±0.06	28.2±0.04	83.5±0.13
6	56.7±2.25	3.3±0.23	27.9±0.57	83.1±0.60
7	56.5±0.17	3.2±0.03	28.1±0.04	83.4±0.05
8	56.6±0.04	3.7±0.03	29.3±0.13	82.9±0.06
9	56.7±0.13	3.2±0.02	27.9±0.05	83.4±0.04
10	58.7±1.68	3.0±0.17	28.4±0.40	83.9±0.43
11	58.3±0.08	3.8±0.10	29.4±0.09	82.6±0.18
12	55.1±0.23	3.7±0.04	28.3±0.06	82.7±0.10
13	60.9±0.63	2.6±0.07	27.9±0.06	84.7±0.15
14	55.8±0.24	3.5±0.04	28.5±0.12	83.0±0.06
15	54.9±0.46	3.6±0.05	28.6±0.06	82.8±0.09
16	61.8±0.21	2.3±0.06	27.7±0.10	85.2±0.12
17	58.1±0.55	3.2±0.01	28.3±0.05	83.6±0.01
18	56.1±0.37	3.5±0.06	28.2±0.04	83.0±0.12

Optimization

Numerical optimization was carried out for the process parameters of spray drying to obtain the best quality

Fig. 3 Superimposed contours for product responses affected by temperature and maltodextrin concentration

Table 4 Regression coefficients of the second order polynomial for the response variables

	DE	a ₀	a ₁	a ₂	a ₃	a ₄	a ₅	r ²	p
Spray drying performance									
Yield (% w.b.)	19	12.6698 (0.9549)	0.1166 (0.9748)	-0.1 (0.9754)	0.01 (0.6557)	2.08E-07 (1.0000)	-0.02 (0.7503)	0.8688	0.1428
	12	1032.573 (0.0375)	-16.4271 (0.0412)	-6.1335 (0.2335)	0.045 (0.2064)	0.0683 (0.0409)	0.0533 (0.5490)	0.9536	0.0325
Properties of powder									
Moisture Content (% w.b.)	19	18.9389 (0.0491)	-0.1764 (0.1681)	-0.1627 (0.1489)	-0.0002 (0.6917)	0.0006 (0.2258)	0.0051 (0.0495)	0.9893	0.0037
	12	6.35 (0.6823)	-0.068 (0.7888)	0.429 (0.1220)	-0.0029 (0.1190)	0.0004 (0.7404)	-0.0042 (0.3556)	0.9461	0.0405
Bulk density (g/ mL)	19	-1.6212 (0.1498)	0.0387 (0.0689)	-0.0413 (0.0413)	0.0003 (0.0522)	-0.0002 (0.0488)	0.0002 (0.0488)	0.9706	0.0167
	12	-1.5229 (0.1456)	0.0289 (0.1103)	0.0165 (0.2341)	-0.0002 (0.1323)	-0.0001 (0.1167)	-6.68E-06 (0.9770)	0.9369	0.0507
Hygroscopicity (g/100 g. d.b.)	19	5.9722 (0.1819)	-0.0796 (0.2567)	-0.0163 (0.7618)	-0.0002 (0.6841)	0.0003 (0.2727)	0.0009 (0.4270)	0.9102	0.0840
	12	7.5489 (0.0020)	-0.1098 (0.0029)	-0.027 (0.0825)	0.0002 (0.1263)	0.0004 (0.0037)	6.67E-05 (0.7629)	0.9966	0.0007
Solubility (s)	19	-95.3297 (0.4643)	2.2833 (0.3123)	1.4667 (0.3655)	-0.01 (0.4324)	-0.01 (0.2908)	3.15E-08 (1.0000)	0.9389	0.0485
	12	123.6698 (0.5523)	-1.43339 (0.6713)	0.83333 (0.7731)	-0.01 (0.6163)	0.005 (0.7200)	0.02 (0.7200)	0.9105	0.0837
Acidity (%)	19	-11.8091 (0.6972)	0.131781 (0.7910)	0.89533 (0.1071)	-0.0033 (0.3044)	-0.00047 (0.8208)	-0.01427 (0.1553)	0.8858	0.1178
	12	25.51437 (0.4547)	-0.41622 (0.4595)	0.383997 (0.4326)	-0.0013 (0.6829)	0.001734 (0.4580)	-0.00627 (0.4985)	0.7054	0.4069
Colour values L*	19	-49.94 (0.6207)	1.4227 (0.4127)	3.4253 (0.0773)	0.0206 (0.1011)	-0.0049 (0.4844)	-0.0248 (0.3924)	0.8688	0.1428
	12	179.1878 (0.2733)	-1.9945 (0.4335)	-1.3650 (0.5264)	0.0105 (0.4776)	0.0079 (0.4517)	0.0205 (0.6160)	0.8973	0.1017
Hue*	19	80.6623 (0.1472)	-0.0415 (0.9555)	0.8457 (0.2484)	-0.0058 (0.2480)	0.0004 (0.8892)	-0.0033 (0.7926)	0.7125	0.3953
	12	87.9970 (0.0025)	1.18E-5 (0.9999)	-1.1833 (0.0029)	0.0048 (0.0128)	-0.0001 (0.8651)	0.0261 (0.0020)	0.9965	0.0007

The bold characters indicate that the corresponding parameter have a significant effect on the response values.

liquorice extract powder. Table 2 shows the independent variables that have a complex relationship with the responses may have more than one maximum point. Analysing the contour plots is the best way to evaluate the relationships between responses and variables. From Fig. 1, they show that the contour plots of yield, moisture content, bulk density, hygroscopicity, solubility, acidity and color index (L and hue value).

The optimum drying conditions can then be determined by superimposing the contour plots of relevant and statistically significant responses. The following limits were proposed: bulk density of not more than 0.6 g/mL, moisture content of at a range of 3–5 %, hygroscopicity of not more than 50 g/100 g, solubility of not more than 30 s, acidity of at a range of 1–2.5 %, L* value of at a range of 55–60, hue* value of not more than 85, and recovery of not less than 50 %. Superimposed contour plots indicated the ranges of variables which could be considered as the optimum range for best product quality as shown in Fig. 3. Superimposing the individual contour plots for the product response variables resulted in the identification of a region (shown by the shaded area) that satisfied all constraints for 12DE (Fig. 3). The optimum ranges of variables obtained from the superimposed contours were 120–130 °C of inlet air temperature and 15–20 % of maltodextrin concentration for 12DE maltodextrin, but it does not have optimum ranges under proposed limits for 19DE maltodextrin. These optimum conditions can be used to produce liquorice extract powder with the estimated characteristics given above.

Conclusions

The liquorice extract powder responses were affected by inlet air temperature, maltodextrin concentration and maltodextrin dextrose equivalent. Addition of maltodextrin reduced the stickiness of powder and increased the yield. The results showed that inlet air temperature has great influence on the physical properties of powder. Moisture content decreases with an increase in inlet air temperature and increase in maltodextrin concentration. Bulk density increases with an increase in dextrose equivalent, a decrease in inlet air temperature and maltodextrin concentration. Hygroscopicity decreases with a decrease in dextrose equivalent, an increase in inlet air temperature and maltodextrin concentration. L* value increases with a decrease in inlet air temperature, an increase in maltodextrin concentration and maltodextrin dextrose equivalent. Increase in temperature, maltodextrin concentration and dextrose equivalent cause a decrease in hue* value. A decrease in the solubility was observed with decreased inlet air temperature (°C), an increased DE maltodextrins, and increased maltodextrin concentration (Table 4).

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