Optimization of bayberry juice spray drying process using response surface methodology

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Abstract In this study, thirteen different runs according to the central composite design-response surface methodology were used to optimize the quality parameters of spray-dried bayberry juice powder. The independent variables were different levels of inlet air temperature (145.8–174.1 °C) and maltodextrin concentration (22.9–37.0%). The responses were process yield, moisture content, hygroscopicity, glass transition temperature (T_g), total color difference (ΔE), redness, retention of phenolics and anthocyanins, and antioxidant capacity. The optimum operation conditions for the highest T_g, redness, anthocyanins retention and antioxidant capacity with the lowest ΔE and hygroscopicity were obtained at inlet drying temperature of 150 °C and maltodextrin concentration of 31%. This study revealed that by applying these optimal conditions, bayberry juice powder with a 74.16% yield, 3.15% moisture content, 10.25% hygroscopicity, 41.15 °C T_g, 24.74 ΔE, 27.45 Hunter *a* redness, 89.55% anthocyanins retention, 77.71% phenolics retention, and 30.19 mmol Trolox equivalents/g antioxidant capacity was produced.

Keywords Anthocyanin · Antioxidant · Bayberry · Glass transition temperature · Spray drying

Introduction

Bayberry, scientifically named *Myrica rubra* Sieb. et Zucc., is a subtropical fruit tree historically cultured in China and other Southeast Asian countries. Bayberry production mainly occurs during May and June. The juicy fruit of bayberry is delicious with attractive color and flavor, and is regarded as a healthy food due to its high vitamin C and other antioxidant phytochemicals contents [1, 2]. However, it is highly perishable and has a short postharvest life. Therefore, bayberries are often processed into juice for more widely available and longer-term consumption.

Conversion of the liquid bayberry juice into a dried fruit powder may further increase the stability of the product and make it easier to transport and distribute. Dried juice powders have long storage life at ordinary temperatures and can be used for making many convenient food products. Spray drying is a common drying technique in pharmaceutical and food industries to convert liquid to powder form. This technique offers short contact time (5–100 s), allowing some heat sensitive ingredients such as flavor, color, and vitamins to be retained in high percentages [3]. Spray drying of fruit juices, however, is problematic since these liquids display thermoplastic behavior due to their high proportion of low molecular weight organic acids and sugars. These components have relatively low glass transition temperature (T_g) and may remain as syrup or stick on the dryer chamber wall, leading to a low product yield and also operating problems. Therefore, several additives including gum, starch, gelatin and maltodextrin are often used as carrier agents to prevent stickiness of product by increasing the T_g of the product during spray drying [4, 5]. Previous study has compared the stickiness overcoming efficiency of protein and maltodextrin on spray drying of

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bayberry juice and found that maltodextrin offered a good compromise between cost and effectiveness [6].

The bayberry juice is found to contain high amount of phenolic compounds and anthocyanins, which not only impart red color to berries, but also exhibit strong antioxidant activities [7]. As phenolics and anthocyanins are sensitive to heat processing [8], spray drying might have the potential to encapsulate and protect bayberry phytochemicals. It is well known that drying processes have great influence on the fruit juice powder quality and physico-chemical properties, such as appearance, texture, porosity, color, and antioxidant capacity [5]. However, until now data on bayberry powders obtained by different drying techniques have been insufficient.

This study aimed to determine optimum spray drying conditions, inlet air temperature and maltodextrin concentration. The spray-dried power was analyzed for process yield, moisture content, hygroscopicity, Tg, total color difference, redness, total phenol and anthocyanin retentions, and antioxidant capacity.

Materials and methods

Raw materials and chemicals

The fully ripened bayberry fruits (cultivar Dongkui) were harvested from a farm located in Shantou, Guangdong Province, China, and transported to the laboratory within 24 h. The fruits were manually cleaned by running cold water to remove any dirt and foreign materials, and then stored at refrigeration temperature (4 °C) for 12 h before juice extraction. Food grade maltodextrin DE 15 was obtained from Xilaike Chem Inc. (Zhengzhou, China). Folin–Ciocalteu's phenol reagent and gallic acid were purchased from Jinshui Biotech (Shanghai, China). ABTS antioxidant assay kit was obtained from Beyotime Biotech (Shanghai, China). All other chemicals were of analytical reagent grade.

Preparation of bayberry juice slurry

The fruits were squeezed using a commercial juice extractor. The juice was sieved through a sieve with 200 meshes to remove pulp and other solids, and then stored at −20 °C until use. The frozen bayberry juice was thawed, and feed solutions were prepared by addition of maltodextrin as a carrier agent. The maltodextrin was first predissolved in a small portion of the juice, and then mixed with the rest of the juice in the blender at a rate of 7500 rpm for 3 min to get uniform slurry. The homogenized juice was spray dried immediately without any delay.

Spray drying

Spray drying process was performed in a pilot-scale spray dryer (Qianjiang Dryer Inc., Changzhou, China). The slurry was kept warm at about 60 °C using a hot-plate magnetic stirrer and stirred at low speed while fed into the spray dryer. The pump rate was adjusted to maintain an outlet temperature of 80 ± 3 °C. Spray-dried bayberry powder was collected. The samples were weighed, packaged using paper foil polyethylene and then stored at 4 °C until further analysis.

Experimental design

Response surface methodology (RSM) was used to derive the optimum formulation conditions (9). A central composite design was applied for two independent variables. The independent variables of different levels of inlet air temperature (X_1) and maltodextrin concentration (X_2) affected the quality of the end product. Table 1 shows the coded and actual levels of experimental design.

A total of 13 experiments in this study were based on two-factor and five levels experimental design, with five replicates at the center of the design to estimate the pure error sum of squares. Dependent variables include process yield (Y_1), moisture content (Y_2), hygroscopicity (Y_3), Tg (Y_4), total color difference (Y_5), redness (Y_6), total phenolics (Y_7) and anthocyanins (Y_8) retention, and antioxidant capacity (Y_9). Multiple regressions were used to analyze the linear, quadratic and interaction terms of independent variables in the response surface models. A second order polynomial of the form of Eq. (1) was assumed to relate the response.

$$Y = \beta_0 + \beta_1 X_1 + \beta_2 X_2 + \beta_{12} X_1 X_2 + \beta_{11} X_1^2 + \beta_{22} X_2^2 \quad (1)$$

where Y is the response; β_0 , β_1 , and β_2 are the regression coefficients for the linear terms; β_{11} and β_{22} are the quadratic terms; β_{12} is the interaction term; and X_1 and X_2 represent the coded values of the independent variables, inlet air temperature and maltodextrin concentration, respectively.

Yield

The spray-drying yield was calculated according to Eq. (2) as followed:

$$\text{Yield (\%)} = \frac{\text{obtained spray dried powder (g)}}{\text{total solids content of juice (°Brix)} \times \text{juice (g)} + \text{maltodextrin (g)}} \times 100 \quad (2)$$

Table 1 Experimental data for response surface analysis of the effects of processing conditions on the quality of bayberry juice powder

Run	Inlet air temperature		Maltodextrin concentration		Yield (%)	Moisture content (%)	Hygroscopicity (%)	Tg (°C)	Color difference (ΔE)	Redness (Hunter <i>a</i>)	Anthocyanins retention (%)	Phenolics retention (%)	Antioxidant capacity (mmol TE g ⁻¹ powder)
	Value (°C)	Code	Value (%)	Code									
1	145.8	-1.414	30	0	69.3 ± 1.8	3.61 ± 0.06	10.1 ± 0.7	40.8 ± 0.5	33.8 ± 4.8	28.0 ± 0.1	87.3 ± 1.3	79.7 ± 3.3	33.4 ± 1.2
2	150	-1	35	1	74.6 ± 2.0	3.14 ± 0.10	9.5 ± 0.0	43.6 ± 1.3	16.5 ± 2.5	26.8 ± 0.2	92.3 ± 0.9	79.9 ± 4.1	28.5 ± 0.4
3	150	-1	25	-1	70.4 ± 1.7	3.30 ± 0.03	11.5 ± 0.8	36.1 ± 0.2	40.5 ± 5.1	27.6 ± 0.9	85.8 ± 1.1	76.2 ± 2.1	37.4 ± 1.5
4	160	0	30	0	72.5 ± 2.1	2.56 ± 0.14	9.7 ± 0.5	41.9 ± 1.4	45.2 ± 7.0	28.0 ± 0.2	82.7 ± 1.0	76.2 ± 2.6	31.7 ± 1.5
5	160	0	30	0	72.9 ± 2.4	2.44 ± 0.14	10.5 ± 0.5	35.2 ± 1.4	19.5 ± 1.2	28.1 ± 0.1	83.3 ± 1.0	76.2 ± 4.5	31.4 ± 1.5
6	160	0	30	0	73.1 ± 2.0	2.54 ± 0.14	10.6 ± 0.5	38.9 ± 1.5	16.5 ± 3.2	28.1 ± 0.2	81.0 ± 1.0	75.1 ± 3.8	31.1 ± 1.5
7	160	0	37.07	1.414	79.2 ± 2.7	2.63 ± 0.05	8.7 ± 1.2	43.9 ± 0.2	14.5 ± 1.8	26.2 ± 0.2	82.4 ± 0.7	86.0 ± 5.5	28.7 ± 1.3
8	160	0	30	0	71.8 ± 2.1	2.81 ± 0.14	10.5 ± 0.5	35.2 ± 1.4	23.4 ± 3.2	27.8 ± 0.2	82.8 ± 1.0	75.1 ± 4.1	31.9 ± 1.5
9	160	0	30	0	73.2 ± 2.2	2.52 ± 0.14	10.5 ± 0.5	36.5 ± 1.4	18.3 ± 1.6	28.0 ± 0.2	83.0 ± 1.0	75.7 ± 3.4	31.1 ± 1.5
10	160	0	22.93	-1.414	60.2 ± 1.5	3.64 ± 0.15	11.0 ± 0.6	33.4 ± 0.8	18.2 ± 1.1	27.9 ± 0.1	78.0 ± 0.9	74.5 ± 2.5	35.0 ± 1.5
11	170	1	35	1	81.2 ± 2.5	2.34 ± 0.04	8.3 ± 0.1	39.6 ± 0.9	55.8 ± 5.6	26.0 ± 0.1	76.1 ± 1.0	81.5 ± 5.3	29.9 ± 2.7
12	170	1	25	-1	65.3 ± 1.1	2.48 ± 0.05	11.7 ± 2.2	35.8 ± 0.2	69.4 ± 8.4	28.0 ± 0.4	73.0 ± 0.3	73.8 ± 3.5	27.9 ± 1.1
13	174.1	1.414	30	0	74.4 ± 1.9	2.51 ± 0.03	10.1 ± 0.2	39.1 ± 0.7	84.3 ± 7.8	27.5 ± 0.1	70.0 ± 0.7	77.5 ± 3.0	28.9 ± 1.4

Determination of moisture content and hygroscopicity

The moisture content spray-dried bayberry powder was determined by the gravimetric method [6]. Hygroscopicity is defined as the mass of moisture (g) absorbed by 100 g of the powder as equilibrium is reached. The experiment was conducted at 25 °C and 75% relative humidity in a desiccator containing sodium chloride saturated solution [9].

Determination of glass transition temperature (Tg)

The Tg of the powder was determined by using Differential Scanning Calorimeter (Model 2010, TA Instruments, Newcastle, USA) with temperature programmer controller. Indium was used for temperature and heat flow calibrations. The samples were scanned from an equilibrium starting temperature of -10 to 120 °C with a heating rate of 10 °C/min. The purge gas used was dry nitrogen (25 mL/min). Thermograms were analyzed using (Thermal Analysis Version 2000) system software.

Color measurements

The color of the juice, feed slurry, powder and reconstituted samples was analyzed using a WR-10 chromameter (Wave Optoelectronics Technology Co., Ltd, Shenzhen, China). 5 g fruit powder was placed into a plastic container with fixed surface to measure the color. In order to compare the color difference before and after spray drying, the powders were converted into juice after reconstitution with known volume of water according to their total soluble solids content. Light beam from the chromameter was passed through the samples and the reflected light was measured by using *L*, *a*, *b* coordinates in the CIELAB system. The instrument was standardized each time with a black and white tile. The color values represented whiteness/darkness (*L*/*-L*), redness/greenness (*a*/*-a*) and yellowness/blueness (*b*/*-b*).

The total color difference (ΔE) was calculated according to Eq. (3):

$$\Delta E = \sqrt{(L_0 - L)^2 + (a_0 - a)^2 + (b_0 - b)^2} \quad (3)$$

where *L*₀, *a*₀ and *b*₀ were initial values of feed mixture and *L*, *a* and *b* are corresponding values of the reconstituted powder.

Anthocyanin determination

Total anthocyanin content (TAC) of juice slurry and powder product was determined by the pH differential

method using two buffer systems: potassium chloride buffer, pH 1.0 (0.025 mol/L), and sodium acetate buffer, pH 4.5 (0.4 mol/L) [10]. Briefly, 0.25 mL of feed solution or reconstituted juice was mixed with 4.75 mL of the corresponding buffers and read against water as a blank at 510 and 700 nm. The total anthocyanin content of each sample (mg cyanidin-3-glucoside/100 mL) was calculated as Eq. (4):

$$\text{TAC} = A \times \text{MW} \times \text{DF} \times 1000 / (\varepsilon \times l) \quad (4)$$

where $A = [(A_{510} - A_{700})_{\text{pH}1.0} - (A_{510} - A_{700})_{\text{pH}4.5}]$, MW is molecular weight (449.2), DF is the dilution factor [20], and ε is the molar absorptivity of cyanidin-3-glucoside (29,600). Anthocyanins retention after spray drying was calculated according to Eq. (5):

$$\begin{aligned} \text{Anthocyanin retention (\%)} \\ &= \frac{\text{TAC in spray dried powder (mg/g)} \times \text{weight (g)}}{\text{TAC in feed solution (mg/mL)} \times \text{volume (mL)}} \\ &\quad \times 100 \end{aligned} \quad (5)$$

Determination of total phenolics

Total phenolics content was determined by the Folin-Ciocalteu method [11]. An aliquot of 0.5 mL of bayberry juice slurry or dissolved bayberry powder solution was mixed with 2.5 mL of Folin-Ciocalteu's reagent (FCR; 1:10 dilution), and incubated at 50 °C for 30 min. 2 mL of Na_2CO_3 (20% solution in water) were added, and the absorbance was measured at 760 nm. Retention of phenolics after spray drying was calculated according to Eq. (6):

$$\begin{aligned} \text{Phenolics retention (\%)} \\ &= \frac{\text{Total phenolics in spray dried powder}}{\text{Total phenolics in feed solution}} \times 100 \end{aligned} \quad (6)$$

Antioxidant capacity determination

Antioxidant capacity was determined by $\text{ABTS}^{\cdot+}$ method according to the procedure as previously described [12]. The $\text{ABTS}^{\cdot+}$ radical cation was generated by reacting ABTS with potassium persulfate. The resulting $\text{ABTS}^{\cdot+}$ solution was maintained in dark for 16 h at 25 °C. An aliquot of 10 μL of different samples was mixed with 200 μL $\text{ABTS}^{\cdot+}$ solution, and incubated at room temperature for 30 min. Then the absorbance of the reaction solution was measured at 710 nm by a SM600 microplate reader (Utrao Medical Instrument Co., Ltd, Shanghai, China), and the scavenging activity on the $\text{ABTS}^{\cdot+}$ free radical was compared with that of the Trolox, a water-soluble vitamin

E analog. Results were expressed in mmol Trolox equivalents (TE)/g of powder.

Statistical analysis

All data are expressed as “mean $\pm$ SD” from at least three replicates. The Design Expert version 8.0 (Statease Inc., Minneapolis, MN) software was used for analysis and response optimization. The level of statistical significance was set at $P < 0.05$.

Results and discussions

Fitted models and response surfaces

Quality parameters of bayberry juice powder under different spray drying conditions are shown in Table 1. Estimated regression coefficients (β) of the quadratic polynomial equation, interactions upon response variables, determination coefficient, and lack-of-fit test and probability are presented in Table 2.

Analysis of variance showed that all response variables could be fit with a second-order polynomial regression model ($P \leq 0.01$). The coefficients of determination values (R^2) obtained for the response variables were more than 0.80, except for Tg (0.72), and these results indicated that a high percentage of response variations could be explained by the response surface models. de Oliveira et al. [13] determined that determination coefficients of higher than 0.80 were good for spray drying of cashew apple juice, and Loh et al. [14] suggested that R^2 values of more than 0.75 were relatively adequate for prediction purposes. The coefficient of variation (CV) of the total color difference was 33.32%, but the CV values of other responses were found within the range of 0.66–7.77%, indicating the experiments had high precision and reliability. The lack-of-fit, which measures the fitness of the models, resulted in a significant P value for only yield, moisture content and phenol retention, and these data suggested that these models were insufficiently accurate for predicting those responses. However, the P values of the models were 0.24 for hygroscopicity, 0.93 for Tg, 0.62 for total color difference, 0.06 for redness, 0.07 for anthocyanin retention, and 0.10 for antioxidant capacity, all of which further indicate the goodness of fit.

Powder yield

Powder yield is a key performance indicator for spray drying process. The high amounts of sugar and organic acid in juice may cause the spray dried product to be sticky,

Table 2 Main effects and interactions of the response surface models in the spray drying of bayberry juice

Coefficients	Yield (Y ₁)	Moisture content (Y ₂)	Hygroscopicity (Y ₃)	Tg (Y ₄)	Color difference (Y ₅)	Redness (Y ₆)	Anthocyanins retention (Y ₇)	Phenolics retention (Y ₈)	Antioxidant capacity (Y ₉)
β_0	72.14	2.53	10.22	38.69	22.80	28.00	82.33	75.69	31.25
β_1	−0.12	−0.38**	−0.11	−0.84	17.44**	−0.15	−6.30**	−0.48	−1.80**
β_2	5.37**	−0.19*	−1.07**	3.27**	−3.81	−0.68**	2.36**	3.46**	−2.47**
β_{12}	3.93*	0.01	Ns	Ns	2.59	−0.28*	−1.61*	0.98	1.73**
β_{11}	Ns	0.17	Ns	Ns	19.53**	−0.18*	−1.41*	1.06*	Ns
β_{22}	Ns	0.27*	Ns	Ns	0.42	−0.57**	−0.65	1.90**	Ns
R^2	0.84	0.86	0.80	0.72	0.85	0.96	0.97	0.93	0.97
Regression (<i>P</i> -value)	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	0.01
Lack of fit (<i>P</i> -value)	<0.01	0.01	0.24	0.93	0.62	0.06	0.07	0.03	0.10
Coefficients of variation (%)	3.38	7.77	4.67	4.92	33.32	0.66	1.70	1.49	1.70

β_0 , constant; β_1 , linear coefficient of inlet temperature; β_2 , linear coefficient of maltodextrin concentration, β_{12} , interaction, β_{11} , quadratic coefficient of inlet temperature, β_{22} , quadratic coefficient of maltodextrin concentration; Ns not significant, $P > 0.05$; * $P \leq 0.05$; ** $P \leq 0.01$

resulting in poor powder recovery [15]. Several researchers studying the spray drying of sticky products, such as those from blackberry and sour cherry juices, considered drying processes with a yield of higher than 50% as efficient [4, 9]. In the present study, the yields of bayberry juice powder varied between 60.2 and 81.2%. The influences of maltodextrin concentration and inlet air temperature on powder yield (Y₁) are shown in Fig. 1(A). The regression coefficients from Eq. (7) indicated that the maltodextrin concentration (X₂) had a linear interaction with inlet temperature (X₁) and was significant on yield. Product yield significantly increased ($P < 0.05$) with an increasing percentage of maltodextrin in the mixture. Maltodextrin encapsulated the organic acids and simple sugars that were present in the bayberry juice and this encapsulation helped the powder particles freely flow through the drying chamber; in other words, product stickiness was successfully prevented. In agreement with the results of the previous study [16], temperature did not have a significant effect on yield ($P > 0.05$).

$$Y_1 = 72.14 - 0.12X_1 + 5.37X_2 + 3.93X_1X_2 \quad (7)$$

Moisture content, hygroscopicity and Tg point

The influences of temperature and maltodextrin on the moisture content (Y₂) of the bayberry powders are shown in Fig. 1(B). The moisture content of powders varied from 2.3 to 3.6%. As shown in Fig. 1(B) and Eq. (8), increasing air inlet temperature or maltodextrin led to lower moisture contents. The addition of maltodextrin to the feed solution

prior to spray drying increased the total solids content and reduced the total amount of water available for evaporation. Previous studies showed that increasing the inlet air temperature increased the drying rate, and, hence, the moisture content of the powder produced [17, 18].

$$Y_2 = 2.53 - 0.38X_1 - 0.19X_2 + 0.01X_1X_2 + 0.17X_1^2 + 0.27X_2^2 \quad (8)$$

The amounts of absorbed moisture in the spray-dried powders after reaching equilibrium are shown in Fig. 1(C). The coefficients of the first order terms for Eq. (9) with coded variables indicated that temperature did not significantly affect the hygroscopicity (Y₃) of the bayberry powder, while maltodextrin did significantly affect hygroscopicity ($P < 0.05$). Increasing the amount of maltodextrin in the feed mixture caused a decrease in the water-holding capacity of powders [15]. Low-molecular-weight components (e.g., simple sugars and organic acids present in fruit juices) have more hydrophilic groups, and therefore, their hygroscopicity is higher. Maltodextrin has a high molecular weight compared to sugars and acids and has a relatively lower hygroscopicity. Therefore, maltodextrin increases the bulk molecular weight of bayberry powder mixture and decreases the water absorption capacity of powders.

$$Y_3 = 10.22 - 0.11X_1 - 1.07X_2 \quad (9)$$

Glass transition temperature can be used to evaluate the processability and storage stability of juice powders. In this study, Tg (Y₄) of spray-dried bayberry powders varied between 35.2 and 43.9 °C. As shown in Fig. 1(D) and

Fig. 1 Response surface plots that display the effects of inlet air temperature and maltodextrin concentration on yield (A), moisture content (B), hygroscopicity (C), Tg (D), total color difference (E) redness (F), retention of anthocyanins (G) retention of phenols (H), and antioxidant capacity (I) of the bayberry powder

Eq. (10), the glass transition temperature of the powder increased with increasing maltodextrin concentration. Fang and Bhandari reported a similar phenomenon: the Tg value increased with increasing amounts of maltodextrin solids because of the higher molecular weight of maltodextrin [6]. Goula and Adamopoulos described that powder hygroscopicity was dependent on the glass transition temperature [19].

$$Y_4 = 38.69 - 0.84X_1 + 3.27X_2 \quad (10)$$

Generally, increasing the maltodextrin concentration during spray drying of bayberry juice resulted in a higher powder glass transition temperature and lower moisture and hygroscopicity values, which may increase the stability of the powder during storage. However, when the maltodextrin concentration was too high, the resulting powder had a lower quality because the nutrients of the fruit were diluted [20].

Color characteristics

Color characteristic is an important indicator of quality; it reflects sensory attractiveness and the quality of the powders produced in the spray drying process. The total color differences (Y_5) of reconstituted powders compared to feed mixtures before the spray-drying process are shown in Fig. 1(E). The regression coefficients of Eq. (11) indicated that a decrease in the inlet air temperature decreased the overall color difference value of the fruit juice powder, while reduction in the maltodextrin concentration of the fruit juice slurry did not show any prominent effect. Moreover, there was no significant relationship between the maltodextrin concentration and drying temperature. Similar results were also observed by other investigators on spray drying of Gac fruit and watermelon [21, 22]. The change in color at high temperatures may be due to the fact that color pigments (e.g. anthocyanin, β -carotene, and lycopene) are sensitive to heat. Another reason may be non-enzymatic browning of simple sugars present in juice [16].

$$Y_5 = 22.80 + 17.44X_1 - 3.81X_2 + 2.59X_1X_2 + 19.53X_1^2 + 0.42X_2^2 \quad (11)$$

The Hunter a value is an important parameter for many juice powders because it is directly related to redness and

product quality. According to the results presented in Fig. 1(F) and Eq. (12), the powder redness (Y_6) had a complex behavior depending on the combination of study factors. However, the obtained Hunter a values exhibited a low variability among the different experiments. The negative linear effect of maltodextrin concentration and also the negative quadratic effect of maltodextrin concentration and drying temperature were significant for powder redness. Results from the optimization process showed that 28% maltodextrin combined with an air inlet temperature of 154 °C led to the highest Hunter a value of bayberry powder. The negative correlation between the air inlet temperature and the percentage of maltodextrin with redness was in agreement with previously reported results found for pomegranate powder [16].

$$Y_6 = 28.00 - 0.15X_1 - 0.68X_2 - 0.28X_1X_2 - 0.18X_1^2 - 0.57X_2^2 \quad (12)$$

In general, the color of the bayberry powder became less red as the maltodextrin concentration and drying temperature increased, both of which must be considered depending on the application of the final product.

Retention of anthocyanins and phenols

Anthocyanins are responsible for the characteristic color of bayberry and its products. When heated, anthocyanins are unstable as they denature or polymerize with other phenolic compounds and thereby lose color [8]. As shown in Fig. 1(G) and Eq. (13), the linear term of inlet air temperature and maltodextrin concentration was statistically significant ($P < 0.01$) for the retention of anthocyanins (Y_7), which varied between approximately 70.0 and 92.3% (Table 2). At low percentages of maltodextrin, the increase in temperature resulted in a sharp decrease in anthocyanin retention. Alternatively, at high maltodextrin concentrations, the increase in temperature resulted in a slight increase in anthocyanin retention. This increase in retention may be due to the high rate of encapsulation of excess maltodextrin present in the feed mixture since encapsulation can prevent anthocyanin degradation. Anthocyanins are thermo-labile, biological materials. As reported by Ersus and Yurdagel [23], higher air inlet (>160–180 °C) and outlet temperatures (107–131 °C) caused a greater loss of anthocyanins after spray drying of black carrot anthocyanins. In the present study, an inlet temperature of 150 °C and outlet temperature of 80 °C were related to a high retention rate of phenols and anthocyanins. Therefore, a high temperature and a high percentage of maltodextrin leads to an efficient spray drying process that protects bayberry phytochemicals from degradation.

$$Y_7 = 82.33 - 6.30X_1 + 2.36X_2 - 1.61X_1X_2 - 1.41X_1^2 - 0.65X_2^2 \quad (13)$$

Bayberry contains abundant phenolic compounds, and they mainly occur in bonded forms, which are more stable than their monomeric forms [24]. The thermal stress of spray drying on the phenolic compounds was expected to be very weak. The phenol retention values ranged from 73.8 to 85.9% [Fig. 1(H)]. These values were remarkably lower than those reported (94%) by Fang and Bhandari [11], which may be due to the relatively high percentage of maltodextrin (50%) used in that study. From the regression coefficients of Eq. (14), it is clear that the maltodextrin concentration had a linear effect ($P < 0.01$) on the retention of phenols (Y_8) while the inlet temperature had a quadratic effect ($P < 0.05$). Higher retention values were obtained at high maltodextrin concentrations and intermediate drying temperatures.

$$Y_8 = 75.69 - 0.48X_1 + 3.46X_2 + 0.98X_1X_2 + 1.06X_1^2 + 1.90X_2^2 \quad (14)$$

Antioxidant capacity

The total antioxidant activities of the powder samples under different spray drying conditions are shown in Fig. 1(I) and Eq. (15). Generally, the two investigated factors, the maltodextrin concentration and the drying temperature, significantly affected the antioxidant capacity (Y_9) of the powders ($P < 0.05$). In a similar pattern as that observed with retention of anthocyanins, increasing the drying temperature resulted in a lower antioxidant activity of powder samples. The lower activity may be explained by the loss of

anthocyanins at higher drying temperatures, and since anthocyanins are a major antioxidant compound in spray-dried bayberry powder, the antioxidant activity diminished. With respect to the influence of maltodextrin concentration, a decrease in the antioxidant capacity was observed when the maltodextrin concentration was increased. This may be due to the dilution effect caused by the addition of maltodextrin, which does not have any antioxidant properties.

$$Y_9 = 31.35 - 1.80X_1 - 2.47X_2 + 1.73X_1X_2 \quad (15)$$

Next, the relationship between the anthocyanin content and antioxidant capacity in the spray-dried powders was investigated [Fig. 2(A)]. The Pearson correlation test showed that there was a significant positive correlation between anthocyanin content and antioxidant capacity ($R = 0.69$, $P < 0.01$), which indicates that by increasing the anthocyanin content, the antioxidant activity in the spray-dried bayberry powders also increased. The increase in antioxidant capacity also agreed well with the observed total phenolic content [$R = 0.83$, $P < 0.01$; Fig. 2(B)]. Natural antioxidants in foods are very effective against free radicals, which negatively affect human health. The high levels of anthocyanins and phenolics with antioxidative activities in the powder make this product highly recommended.

Optimization and validation

The powders can be used for various purposes such as a reconstituted fruit juice after dissolving in water or as a food additive for improving the taste, color, or nutritional value of solid beverages, desserts, and other food and nutraceutical products. The desired product characteristics were considered as follows: low hygroscopicity, high Tg, high anthocyanin retention, and good preservation of

Fig. 2 Correlation of the antioxidant capacity with the contents of anthocyanins (A) and phenols (B) in the bayberry powder

Fig. 3 Response surface plots showing the effects of inlet air temperature and maltodextrin concentration on the overall response desirability of spray-dried bayberry powder

Table 3 Response variables for the optimum conditions for obtaining powder from bayberry juice

Variables	Goal	Theoretical values	Experimental values
Yield (%)	In range	72.2 ± 2.2	74.2 ± 3.5
Moisture content (%)	In range	3.03 ± 0.14	3.15 ± 0.17
Hygroscopicity (%)	Minimize	10.1 ± 0.5	10.2 ± 0.9
Tg (°C)	Maximize	40.4 ± 1.9	41.1 ± 1.2
Color difference (ΔE)	Minimize	23.3 ± 11.7	24.7 ± 6.2
Redness (Hunter <i>a</i>)	Maximize	27.8 ± 0.2	27.4 ± 0.2
Anthocyanins retention (%)	Maximize	88.2 ± 1.4	89.6 ± 2.1
Phenolics retention (%)	In range	78.0 ± 1.2	77.7 ± 3.3
Antioxidant capacity (mmol TE/g powder)	Maximize	32.1 ± 0.5	30.2 ± 1.8

powder color and antioxidant capacity. To determine the optimum spray-drying process conditions with desirable response goals, a desirability function was used for numerical optimization (Fig. 3). As a result, the predicted optimum temperature and percentage of maltodextrin were 150 °C and 31%, respectively.

In order to check the adequacy of the response surface model, three experiments were performed under the recommended optimum conditions. The experimental values were consistent with the predicted values (Table 3). Furthermore, theoretical validation using the two sample *t* test on the experimental data and fitted data showed that there was no significant difference ($P > 0.05$) between those values. The optimal properties of spray-dried powder obtained from this study were 74.16% of yield, 3.15% of

moisture content, 10.25% of hygroscopicity, 41.15°C of Tg, 24.74 of ΔE , 27.45 Hunter *a* of redness, 89.55% of anthocyanins retention, 77.71% of phenolics retention, and 30.19 mmol TE/g of antioxidant capacity. Overall, the results obtained in this study indicate that good quality bayberry powders with high anthocyanin and phenolics retention can be produced by spray drying at inlet temperature of 150°C and adding maltodextrin concentration at 31%.

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Compliance with ethical standards

Conflict of interest The authors declare no conflict of interest.

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