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ORIGINAL ARTICLE

Development of a food colorant from *Syzygium cumini* L. (Skeels) by spray drying

Fabián M. Gaibor[1] · Daliannis Rodríguez[2] · Mario A. García[3] · Carlos M. Peraza[2] · Danay Vidal[2] · Antonio Nogueira[4] · Alicia Casariego[2]

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Abstract The objective of the present work was to develop a powder colorant for food use by spray drying from a hydroalcoholic extract of black cherry (*Syzygium cumini* [L.] Skeels). The content of total solids significantly affected the contents of anthocyanins and total polyphenols, while the air inlet temperature influenced ($p \leq 0.05$) the spray drying performance. The optimal drying conditions were 165 °C as air inlet temperature and 25% of total solids, which allowed obtaining a powder colorant with total anthocyanin contents between 4273 and 5070 mg/1000 g, total polyphenols from 10,142 to 11,184 mg/1000 g, and a drying yield between 67.14 and 67.7%. The colorant presented 5.65% humidity, 25.2% hygroscopicity, poor fluidity, and high cohesiveness, with a dissolution time of 55 s. The degradation of anthocyanins, adjusted to zero-order kinetics, was directly proportional to the increase in temperature and time. The values of the component a* decreased with increasing temperature and time.

Keywords *Syzygium cumini* · Anthocyanins · Spray drying · Powdered colorant · Physical properties · Degradation of anthocyanins

✉ Mario A. García
marioifal@gmail.com

[1] Universidad de Investigación de Tecnología Experimental Yachay, Imbabura, Ecuador

[2] Pharmacy and Food Institute, University of Havana, St. 222 No. 2317, 13600 Havana, ZC, Cuba

[3] Biological Sciences Department, Faculty of Health Sciences, Manabi Technical University, Ave. Urbina and Che Guevara, Portoviejo, Manabí, Ecuador

[4] Drugs Research and Development Center, Ave. 26 No. 1605., Havana, Cuba

Introduction

Color is a quality characteristic of food products and plays a fundamental role because it is associated with a taste threshold, a sweet perception, food preference, and acceptability. According to Downham and Collins (2000) there are four types of colorants available in the food market: synthetic (42%), natural (27%), identical natural (20%), and caramel (10%). The use of synthetic colorants is based on the fact that they are cheaper, stable, and brighter than natural colorants. However, in various studies, some of them have been related to carcinogenesis, genotoxicity, and neurotoxicity (Merinas-Amo et al. 2019).

For their part, natural colorants are obtained from inexhaustible sources: plant material, insects, algae, cyanobacteria, and fungi. The most recent consumer trends demand natural products, with therapeutic and medicinal properties, among others, due to the toxicity attributed to synthetic colorants.

Anthocyanins are pigments responsible for a wide range of colors ranging from red to purple in flowers, fruits, leaves, stems, and roots of plants (Castañeda et al. 2009). These pigments are flavonoids with antioxidant activity that have been linked to the prevention of cardiovascular and neurological diseases, cancer, and diabetes (Konczak and Zhang 2004).

The black cherry (*Syzygium cumini* [L.] Skeels) is a fruit of an intense purple color recognized in India, country of origin, for its nutritional and physiological benefits. Several studies have reported the high anthocyanins content of this fruit (Gaibor et al. 2016), comparable to that of blueberries, considered a nutraceutical product of high commercial value. That is why the black cherry has been an interesting subject in the research related to the development of natural colorants rich in anthocyanins. In this context, Gomez et al. (2016) evaluated the biological activities of an aqueous extract from black cherry atomized by spray drying. Shwetha and Preetha (2017) compared the freeze-drying and spray drying of an aqueous extract from black cherry. In addition, it was studied anthocyanin's stability in these powders (Shwetha and Preetha 2016). In addition, Singh et al. (2019) developed a powder from the spray drying of black cherry pulp.

Although spray drying is the most frequently used method in the food industry, it often results in the degradation of the phytochemicals present in the product to be processed. For this reason, proper drying techniques must be employed to preserve the original components and minimize the detrimental effects of drying. In this sense, the incidence of the air inlet temperature and total solids content in the spray drying of a hydroalcoholic extract of black cherry was evaluated to obtain a stable colorant with potential use in the food industry.

Materials and methods

Selection and harvest of black cherry fruits

Ripe black cherry fruits were harvested taking into account similarities in size, color, uniform ripeness, and the absence of visible defects. The pulp and skin were ground in a digital Ultra-Turrax IKA T25 (model T25 D S25).

Preparation of the hydroalcoholic extract of black cherry

The extraction of anthocyanins from the pulp was carried out by maceration with ethanol at 90% (v/v) acidified with 0.03% (w/v) of citric acid and a ratio of 1 g per 5 mL. The mixture was kept, at room temperature, for six hours on a sieve (Retomed, Mizard. 2001) at 285 min^{-1}. After this time, the solid residue was filtered and discarded (Gaibor et al. 2017). This diluted hydroalcoholic extract was determined the pH and extraction yield of polyphenols and total anthocyanins. Both yields (%) were calculated as the ratio between the contents of the extract and the pulp.

Obtaining the concentrated extract

The diluted hydroalcoholic extract was concentrated to a total solids content between 14 and 15% in a rotary evaporator (IKA, Germany) at 40 °C with a rotation of 85 min^{-1}, vacuum pump (ULVAC DTC-21). In addition, the concentrated extract was determined its density with a pycnometer (Mercali et al. 2011), refractometric soluble solids, and total solids in thermogravimetric balance (Sartorius, Göttingen, Germany) until constant mass at 105 °C. The determinations were made in triplicate and the mean value was reported. In addition, the contents of total polyphenols and anthocyanins were determined (Gaibor et al., 2016). The content of total polyphenols was expressed as gallic acid in mg/100 g of pulp and that of anthocyanins as cyanidin-3-glucoside.

Spray drying optimization

Numerical optimization was performed through an Optimal IV response surface design using the Design Expert 8.0.6 program (Stad-Ease Inc., Minneapolis, USA). The optimized factors were the air inlet temperature (A) and total solids content (B); while, the response variables were the retention of total anthocyanins, polyphenols, and drying performance.

The experimental matrix for spray drying the black cherry extract yielded 8 runs including three replicates. The air inlet temperature values were 150, 165, and 180 °C, while those of total solids were 25 and 30% (w/w) in correspondence with Arrazola et al. (2014). The selection of the interval was related to adding the least amount of maltodextrin (11 and 16%) to reduce stickiness and obtain satisfactory yields. In addition, to the extent that the amount of maltodextrin was increased to increase the total solids of the mixture, the coloring power and biological activity of the powder would be affected.

Spray drying

Maltodextrin DE 12 was added to 50 mL of the concentrated extract for a final content of total solids according to the design specifications for each run. The drying was carried out in a spray dryer Mod. B-191 (BÜCHI, Switzerland) with a 0.7 mm diameter nozzle and 60 m^{3}h^{-1} as airflow when the aspirator is at 100%. The air inlet temperature was guaranteed by preheating the equipment for 15 min. The outlet temperature was kept constant at 70 °C, the feed flow was 46% (678 mL/h) for 150 °C, 65% (963 mL/h) for 165 °C and 76% (1128 mL/h) for 180 °C. The determinations were made in triplicate and the mean value was reported. The yield of the drying process was considered as the ratio between the mass of the powdered colorant and the total mass of the total solids of the feed.

Characterization of the powdered colorant

Retention of total polyphenols and anthocyanins

The retention of total polyphenols and anthocyanins (% w/w) was determined by the relationship between the content of these compounds in the powder and their content in the feed for drying.

Hygroscopicity

Powdered colorant (1 g) was placed in a desiccator with a saturated sodium chloride solution to generate a relative humidity of 75.29% at 25 °C. To determine hygroscopicity, the final mass after one week and the initial humidity of the powdered colorant were recorded. It was expressed as water in g/100 g of dry solids (Tonon et al. 2009).

Dissolution time

To 1 g of powdered colorant, in a beaker, 25 mL of distilled water were added at 25 °C, it was put under magnetic stirring at 900 min^{-1} and with a stirring bar of 8 × 22 mm. The dissolution time was taken as the time required to completely dissolve the colorant (Goula and Adamopoulos 2010).

Density

Bulk density

The volume that took up 0.5 g of powdered colorant was recorded in a test tube to calculate the bulk density as the ratio of the mass to the volume of the sample (Shah et al. 1997).

Compacted density

The volume occupied by 0.5 g of the powder colorant in a cylinder after continuously beating it was measured (Shah et al. 1997). Compacted density refers to the ratio of the mass of the sample to the volume of the compacted sample.

Particle density

Powdered colorant (0.5 g) was added in a 10 mL volumetric flask, it was made up with toluene and the volume consumed was recorded until reaching the gauging (Shah et al. 1997). The particle density was obtained from the ratio of the mass of the sample between the difference of the total volume of the volumetric flask and the consumed volume of toluene.

Powder fluidity

The powder fluidity was expressed as Carr's index (CI) or compressibility and was calculated using Eq. 1 (Jinapong et al. 2008).

$$CI = \frac{D_c - D_a}{D_c} \times 100 \quad (1)$$

where: CI, Carr index; Da, bulk density; Dc, compacted density.

The classifications of the fluidity of a powder according to the value of the Carr index would be very good for values less than 15; good, between 15 and 20; acceptable, between 20 and 35; bad, between 35 and 45; and very bad for values greater than 45.

Hausner index

The Hausner index, a measure of the flow properties of powders, was determined as the relationship between compacted density and bulk density. Values lower than 1.25 correspond to free-flowing powders, while higher values correspond to poor flow capacity (Hayes 1987).

Angle of repose

The angle of repose was determined by the funnel method. 10 g of powdered colorant were weighed out and placed in the funnel set at 20 cm in height. The sample was allowed to flow freely and the diameter and height of the formed cone were measured. The angle of repose was obtained by Eq. 2 (Cooper and Gunn 1986).

$$\theta = \tan^{-1}\left(\frac{h}{r}\right) \quad (2)$$

where: h, the height of the cone; r, the radius of the cone.

According to Barbosa-Cánovas et al. (2005), a powder with values lower than 35° is considered free of flow, between 35 and 45° it is classified as quite cohesive, from 45 to 55° as cohesive, and values higher than 55° as very cohesive.

Coloring power

The coloring power was analyzed through the values of the component a* and it was determined at a colorant solution at 0.04 g/mL with the use of a UV–VIS spectrophotometer (Rayleigh UV-1601, Beijing) according to the procedure of Ruiz et al. (2017).

Stability of powder colorant

Approximately 3 g of the powdered colorant were packed in multilayer bags (aluminum and polypropylene). They were placed in a stove Mod. YLD-6000 (AISET, China), at 45, 50 and 55 °C. The stability of the colorant was determined from the estimation of the degradation kinetics of total anthocyanins and the variation of the red-green component (a*). The Q_{10} factor was also estimated according to Eq. 3.

$$Q_{10} = kT + 10/kT \quad (3)$$

where: k, the rate constant of the reaction; T, temperature (°C).

The kinetic parameters (reaction order and rate constant of the reaction) of anthocyanin loss and variation of component a* during accelerated storage of black cherry powder colorant were estimated through the graphical path of the integral method. The integral method consists in assuming the form of the differential velocity equation with orders (n) defined. The integration of this formation will allow deducing a functional relationship between concentration and time and will introduce the rate constants (k) as parameters to be determined by fitting the integrated rate formation chosen by the researcher to the experimental measurements.

Statistic analysis

Analysis of variance was performed and, if there were significant differences, Duncan's multiple range test was applied to determine the differences with a confidence level of 5%. The Statistics program (version 7, 2004, StatSoft. Inc., Tulsa, USA) was used.

Results and discussion

Characterization of the concentrated extract

The black cherry extract concentrated by rotary evaporation, before being mixed with the maltodextrin DE 12, presented mean values of 66.91 mg / 100 g of total anthocyanins, 1781.32 mg/100 g of total polyphenols, 14.64% of total solids, 20.73 °Brix and 1.1033 g/mL density. These results generally corresponded to those of Silva et al. (2013) for jaboticaba extracts, with mean values of 60.57 mg/100 g of total anthocyanins, 82.59% humidity, 17.41% of total solids, and 20 °Brix.

Regression models and statistical analysis

The relationships between the retention of anthocyanins and total polyphenols were analyzed, as well as the performance of the drying process with the independent variables air inlet temperature and total solids content. Table 1 shows the experimental design and results for each variable.

As can be seen, the highest retention of anthocyanins and total polyphenols was obtained for 180 °C with 25% of total solids, corresponding to run 2; however, it presented one of the lowest yields in drying. On the other hand, the highest drying performance was presented by run 4, with 30% (w/w) of total solids and at 165 °C.

The replicas of the study, runs 1, 3, and 4, differed significantly in the retention of total anthocyanins; not so for the retention of total polyphenols. In the case of drying performance, the three runs presented the highest yields.

The responses obtained for each of the runs were related to the two independent variables through linear mathematical equations for the retention of anthocyanins and total polyphenols and quadratic for the drying performance. The best fit of the variables to the models was:

$$\text{TAR} = 68.97 + 0.78\text{A} - 7.97\text{B} \quad (4)$$

$$\text{TPR} = 61.60 + 1.14\text{A} - 7.08\text{B} \quad (5)$$

$$\text{SDP} = 63.42 - 5.09\text{A} - 1.62\text{B} - 1.60\text{AB} - 26.38\text{A}^2 \quad (6)$$

where: TAR; retention of total anthocyanins; TPR, total polyphenol retention; SDP, spray drying performance; A, inlet air temperature; B, percentage of total solids.

Table 2 shows the analysis of variance and significance of the estimated coefficients for each model as a function of the response variables. The degree of fit of the experimental data to the suggested models was evaluated using the parameters F, p, R^2, and lack of fit.

In the case of the retention of anthocyanins and total polyphenols, it was observed that the F values of the suggested models were significant; therefore, the linear models described, with a 95% confidence level, the relationship between the independent variables and the retention of anthocyanins and total polyphenols. For the drying performance, the quadratic model was the one that best adjusted the experimental results, for the same level of confidence.

The percentage of total solids was the factor that significantly affected the retention of anthocyanins and total polyphenols (Table 2), which could be related to the protective effect of maltodextrin during the spray drying. In addition, the maltodextrin prevented stickiness of the product, due to the presence of sugars and low molecular weight acids, which have a low glass transition temperature (Tg), which could decrease the yield of the process. The drying performance was influenced by the linear and quadratic terms of the inlet temperature, this latter with greater significance.

In addition, the coefficients of determination (R^2) were evaluated as an indicator of the fit of the models. In an experimental design, R^2 is an indicator of the variation around the mean, explained by the model. This coefficient increases whenever significant factors are added to the model, while the adjusted R^2 value does not. In all cases, the value of the R^2 statistic indicated that the adjusted

Table 1 Effect of the inlet temperature and total solids on the retention of total anthocyanins, polyphenols and drying performance

Run	Inlet temperature (°C)	Total solids (% w/w)	Retention of total anthocyanins (%)	Retention of total polyphenols (%)	Drying performance (% w/w)*
1	165	30	65.20 (0.02) c	56.9 (0.3) b	60 (2) b
2	180	25	79.05 (0.02) a	71.4 (0.3) a	35 (1) e
3	165	30	58.369 (0.003) f	53.9 (0.2) b	60.6 (0.9) b
4	165	30	60.209 (0.009) e	56.4 (0.1) b	65 (1) a
5	150	30	61.14 (0.02) d	53.6 (0.2) b	42.1 (0.7) cd
6	180	30	60.05 (0.01) e	53 (1) b	28.7 (0.3) f
7	150	25	75.27 (0.03) b	67.3 (0.3) a	40 (1) d
8	150	25	75.71 (0.02) b	66.3 (0.2) a	44.2 (0.1) c

Mean (Standard deviation); n = 3

Different letters indicate significant differences at $p \leq 0.05$

*Expressed on a dry basis

Table 2 Analysis of variance of the models for the retention of total anthocyanins, polyphenols and drying performance

Source	Retention of total anthocyanins		Retention of total polyphenols		Drying performance	
	F value	p value	F value	p value	F value	p value
Model	36.51	0.0010	50.56	0.0005	34.80	0.0076
A	0.45	0.5325	1.70	0.2495	13.70	0.0342
B	71.80	0.0004	100.50	0.0002	1.38	0.3241
AB	–	–	–	–	1.36	0.3277
A^2	–	–	–	–	96.70	0.0022
R^2	0.9359	–	0.9529	–	0.9789	–
R^2 adjusted	0.9103	–	0.9340	–	0.9508	–
Lack of fit	0.40	0.7013	4.90	0.1134	–	–

A: Inlet temperature; B: percentage of total solids

models explained more than 90.0% of the variability of the response variables.

The distribution of the residuals allowed evaluating the adequacy of the models by determining that the residuals followed a normal distribution. The residuals are the differences between the predicted and actual values, which are also expected to have a normal distribution. The adjustment of the studentized residuals, in each case, to a straight line, indicated that they followed a normal distribution. When the data do not meet a normal distribution, an S-shaped curve is generally formed, and the generated models tend to be inaccurate (Chauhan and Patil 2013). The results indicated that the suggested models were appropriate.

Influence of inlet air temperature and total solids content on total anthocyanin retention

Statistical analysis identified the percentage of total solids as the variable that significantly affected anthocyanin retention. It is observed in Eq. 4 and Table 2 that the effect of this factor was linear and negative, that is, with its increase, anthocyanin retention decreased. This behavior could be related to the addition of maltodextrin to reach the required total solids content; the lower the total solids content, the lower the addition of maltodextrin, and the higher the anthocyanin content.

Although the air inlet temperature did not influence significantly, with its increase, the total anthocyanin retention increased. The maximum retention of anthocyanins was obtained between 170 and 180 °C, and 25 and 26% of total solids (Fig. 1a). Silva et al. (2013), who informed similar results through statistical convenience, obtained the best anthocyanin retention with 30% maltodextrin at 180 °C. In addition, Arrazola et al. (2014) for drying anthocyanins extracted from the peel of eggplant (*Solanum melongena* L.) used 180 °C as air inlet temperature and 30% of total solids with maltodextrin as encapsulant agent. The stability of the anthocyanins is affected by various factors such as pH, temperature, concentration, light, oxygen, among others, and it cannot be categorically affirmed that the increase in the air inlet temperature

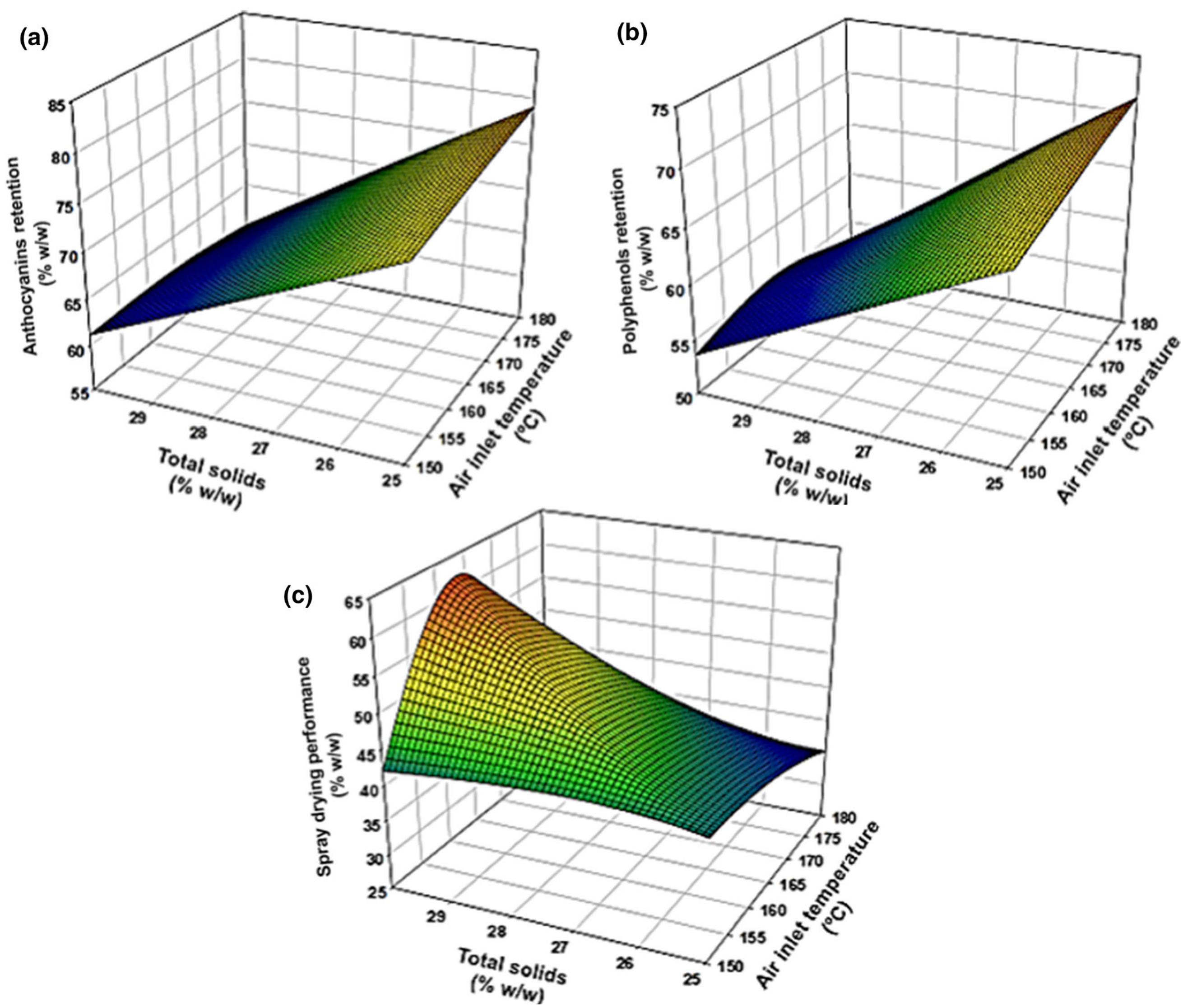

Fig. 1 Influence of air inlet temperature and total solids content on **a** anthocyanins retention; **b** polyphenols retention; and **c** drying performance

produced an increase in the value of this response variable, and the increase could be related to a fluctuation in the measurement process.

Similarly, Bakowska-Barczak and Kolodziejczyk (2011) reported that temperatures below 180 °C did not significantly affect the anthocyanin content of powders obtained by spray drying of an alcoholic extract of black currant berries (*Ribes nigrum* L.). However, Santhalakshmy et al. (2015) concluded that air inlet temperatures, higher than 150 °C in the drying of black cherry juice mainly affect the color of the powder, in addition to the physical properties, taking this temperature as optimal.

In general, there is a consensus on the use of air inlet temperatures of up to 180 °C in the spray drying processes of plant extracts, related mainly to the physical properties and biological activity of the powdered products that are obtained by this drying technique.

Influence of the air inlet temperature and total solids content on the retention of total polyphenols

Table 2 shows that the air inlet temperature was not significant ($p > 0.05$), while the percentage of total solids was. Although this factor was not significant in the model, it existed, as a trend on the content of total polyphenols, the same as on the retention of anthocyanins, linear and negative (Eq. 5). It is observed in Fig. 1b that the highest retention values of total polyphenols, as well as for the anthocyanin content, were obtained for air inlet temperatures between 170 and 180 °C and a percentage of total solids between 25 and 26%.

As in this work, according to Arrazola et al. (2014), the air inlet temperature (170 to 180 °C) did not affect the polyphenol content. On the other hand, Bakowska-Barczak and Kolodziejczyk (2011) reported that the total polyphenol content behaved the same as that of black currant anthocyanins and was not affected by the dextrose equivalent of maltodextrin.

Influence of air inlet temperature and total solids content on drying performance

The linear and quadratic terms of the inlet temperature for the case of drying performance were significant (Table 2). The negative signal in the quadratic term (Eq. 6) corresponds to a process with a maximum, as can be observed in Fig. 1c as a curvature around 160–165 °C.

Bakowska-Barczak and Kolodziejczyk (2011) achieved the highest yield of 86% with maltodextrin DE 11 at 150 °C, although they did not report the total solids content for a better comparison, and the extract to be encapsulated was adjusted to 35 °Brix. Arrazola et al. (2014) obtained the highest yield (90.74%) in drying eggplant anthocyanins at the same percentage of total solids as in this study and using maltodextrin, but for an air inlet temperature of 180 °C. The performance was significantly affected, both by the inlet temperature and by the concentration of the encapsulating agent, not so in the present work.

A similar yield (96.15%), but higher than that of Arrazola et al. (2014), was reported by Begum and Deka (2017) in the drying of an extract rich in anthocyanins at 170 °C inlet temperature, 20 °Brix and with the addition of maltodextrin.

Optimization of drying parameters and verification of models

The retention of anthocyanins and total polyphenols, as well as the higher performance in the drying process, were the variables to consider. In this sense, their values were maximized by maintaining the intervals of the independent variables. Statistical convenience was used to select the optimal process conditions. To verify the models, three dryings were carried out at the optimal conditions (air inlet temperature of 165 °C and 25% of total solids) with the highest statistical convenience (0.915).

The predicted values from the numerical optimization of the process corresponded to 76.9, 68.7, and 68.9% for TAR, TPR, and SDP, respectively; while TAR and TPR at optimal experimental conditions were lower, with mean values (standard deviation) of 68 (6) and 61 (3) %, respectively. The yield under these conditions was 67.6 (0.4) %.

The greater retention of anthocyanins and total polyphenols predicted by the models concerning those obtained experimentally could be related to external factors that weaken the chemical stability of these compounds. In addition, to verify the results, different batches of fruit were used and, therefore, the use of the chemical composition of the fruits used at each moment of the investigation would influence. Thus, it must be taken into account that the predictive values constitute an approximation to this inherent intrinsic value caused by the use of raw material with variable composition.

Bakowska-Barczak and Kolodziejczyk (2011) reported, for colorants dried with maltodextrins, anthocyanin contents (361 to 454 mg/100 g) and total polyphenols (914 to 1251 mg/100 g), similar values to those obtained in this study. However, Begum and Deka (2017) reported anthocyanin values (57.29 mg/100 g) lower than those of the present study for a colorant obtained at 170 °C and 20% of total solids. The drying yields of hydroalcoholic extracts rich in anthocyanins are highly variable, ranging between 8.25 (Santhalakshmy et al. 2015) and 96% (Arrazola et al. 2014) and are influenced by biological factors of the plant material (type cultivar and degree of maturity), climatic conditions, extraction method, air inlet temperature, percentage of total solids and type of encapsulant. On the other hand, through the optimization of the spray drying of a black cherry pulp, 185 °C as the air inlet temperature and 10% maltodextrin were acquired as the best process conditions (Singh and Paswan 2019).

Characterization of black cherry powder colorant

The results of the different physical indicators that were determined to the black cherry powder colorant obtained with the optimized parameters are presented in Table 3. Moisture content is an important property of powdered products and is related to the efficiency of the drying.

The moisture content of the colorant was 5.65%, similar to the 5.72% reported by Begum and Deka (2017). Both values were higher than those reported by Santhalakshmy et al. (2015) from 3.22 to 4.18% for powders obtained from

Table 3 Physical and chemical properties of the black cherry powder colorant obtained with the optimized parameters of spray drying

Parameter	Mean (Standard deviation)
Humidity (% w/w)	5.65 (0.03)
Dissolution time (s)	55 (1)
Angle of repose (°)	46 (2)
Powder fluidity (%)	44 (2)
Cohesiveness	1.73 (0.19)
Bulk density (g/mL)	0.2389 (0.0008)
Compacted density (g/mL)	0.411 (0.004)
Particle density (g/mL)	0.63 (0.01)
Hygroscopicity (g/100 g b.s.)*	25.2 (0.5)

* Determined at 504 h

black cherry juice. The differences in moisture content between these studies could be related to the air inlet temperature used in drying, since these authors stated that, with the increase in this factor, the value of the response variable increased.

The hygroscopicity of the colorant at one week (168 h) was 25.2%; similar to 25.33 and 25.35% reported by Santhalakshmy et al. (2015) and Arrazola et al. (2014) for air inlet temperatures at 160 and 170 °C, respectively. A close, but a higher value (28.89%), was reported by Begum and Deka (2017) for the same study times and type of fruit.

Fluidity (Carr's index) and cohesiveness (Hausner's index) are properties of powdered products that are related to each other, since the higher the cohesiveness, the lower the fluidity. The fluidity of the colorant was 44% and the Hausner index was 1.73; According to both indicators, the powder presented poor fluidity, so the black cherry colorant was characterized by being highly cohesive. The cohesion of powders determines their consistency and flow properties; thus, a lower cohesion will allow a better fluidity of the powders (Domian and Poszytek 2005).

Values close to those found in this work for fluidity (36.10%) and cohesiveness (1.57), presented a powdered colorant obtained from black cherry juice (Santhalakshmy et al. 2015). These authors also demonstrated the influence of the air inlet temperature on these parameters. As this factor increased, both indices decreased.

The angle of repose is that between the inclined surface and the base; provided that the base remains stable, without slipping (Buitrago et al. 2004). The value of the angle of repose of the colorant (46°) confirms the cohesiveness of the powder concerning the classification reported by Barbosa-Cánovas et al. (2005).

The bulk density of the powder colorant was 0.2389 g/mL, a value similar to that reported by Santhalakshmy et al. (2015) of 0.25 g/mL. The inlet temperature did not affect this property. The lower the bulk density, the more air is trapped within the powders, which translates into a greater possibility of product oxidation and less stability during storage. The compacted density of the colorant was 0.411 g/mL. Particle density measures the volume of the pores in the powder; the black cherry stain presented a value of 0.63 g/mL for this parameter.

One of the fundamental properties of powdered products is their solubility because, among other reasons, it allows defining the field of application. The dissolution time of the powdered colorant in water was 55 s, so the powder is highly soluble in water and this capacity allows it to be used in the food industry.

Evaluation of the stability of the powdered colorant during accelerated storage

Loss of total anthocyanins

The loss of anthocyanins from the black cherry powder colorant during accelerated storage was directly proportional to the increase in temperature and time. As seen in Fig. 2a, at the end of storage, about 40% had been lost; 60 and 80% of total anthocyanins at temperatures of 45, 50, and 55 °C, respectively.

In this sense, Begum and Deka (2017) reported a gradual decrease of 44% in the anthocyanin content for day 21 of storage at 30 °C. The degradation of anthocyanins reported by these authors was higher than the 20% obtained in this work for the same storage time at 45 °C. On the other hand, Bakowska-Barczak and Kolodziejczyk (2011) reported that the total anthocyanin content of encapsulated powders was reduced by 35% during 12 months of storage at 8 °C, while at 25 °C, the reduction was 32%.

The degradation kinetics of anthocyanins was monitored during the storage period. The experimental data were adjusted to a zero-order reaction, with R^2 values of 0.9558, 0.9773, and 0.9331 for 45, 50, and 55 °C, respectively. The

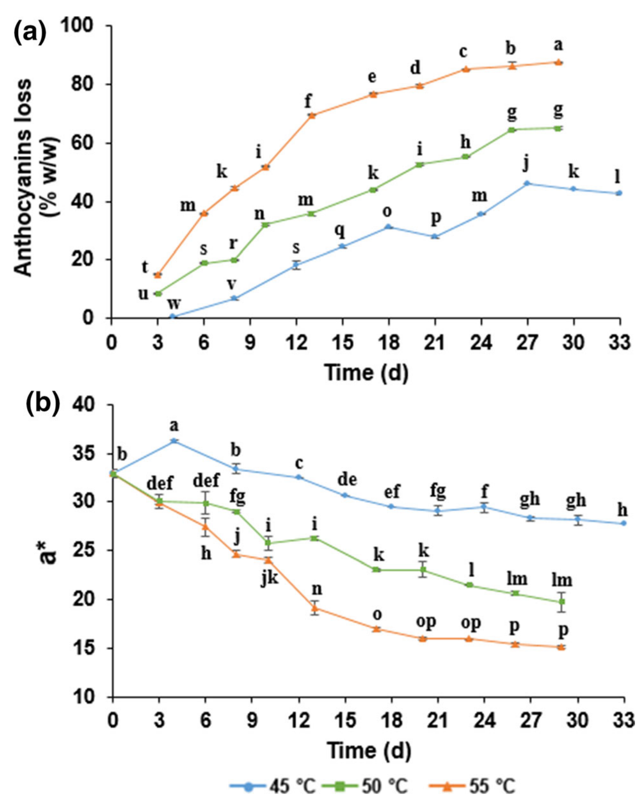

Fig. 2 Anthocyanins losses (**a**) and changes in the component a* (**b**) in the black cherry powder colorant during its accelerated storage. Error bars represent the standard deviation. Different letters indicate significant differences at $p \leq 0.05$

increase in the values of the speed constant was proportional to the temperature, for which there was greater degradation of the pigment at 55 °C. In general, it has been suggested that an increase in storage temperature conditions an increase in speed constants (Table 4).

These rate constants can be used to estimate the value of Q_{10} as a measure of the sensitivity of reactions to temperature. This coefficient informs the increase in the speed of a reaction when the temperature varies by 10 °C. The loss of anthocyanins in the black cherry powder colorant presented a Q_{10} value of 1.54, determined as the relationship between the rate constants at 55 and 45 °C.

Contrary to the results of this study, it has been reported in other studies that the loss of anthocyanins corresponded to first-order degradation kinetics (Arrazola et al. 2014). The higher rate of degradation of anthocyanins can be attributed to the non-encapsulated material, which therefore has more contact with oxygen. In addition, greater adsorption of water at the beginning of storage can also influence by facilitating molecular mobility, related to the effect of water activity on the degradation of anthocyanins.

Variation of component a*

The values of the component a* (red-green) decreased within the positive region, that is, the color varied from a bright reddish tone to brownish, opaque, and brownish tones. It is observed that the component a* varied significantly with temperature, so that the degradation of anthocyanins, responsible for the red color, was much faster at the higher temperature, with a reaction rate of 0.0126 d^{-1}. On the other hand, after 12 days of storage at 45 °C, the variation was minimal. Similar results were obtained by Begum and Deka (2017) with a gradual decrease in the red color of a natural colorant in powder during its storage, as well as an increase in the value of the component b*, translated into an increase in the yellow color. Variations in component a* could be attributed to the degradation of anthocyanins during storage.

The experimental data of the variation of the values of the component a* were adjusted to first-order kinetics, with values of the coefficient of determination of 0.9674, 0.9869, and 0.9485 for 45, 50, and 55 °C (Table 4). Similar to this work, Casati et al. (2015) reported that the reduction of the component a* at 40 °C was adjusted to a first-order model, with a k-value of 0.0064 d^{-1}. According to this value, the degradation at 40 °C of the anthocyanins in this matrix was faster than the degradation at 45 °C in this study. The activation energy values for the thermal degradation of anthocyanins and component a* (144.6 and 140.5 $kJmol^{-1}$) indicated the temperature dependence of these degradation processes. The variation of the values of the component a* presented a Q_{10} value of 5.48, which is related to the fact of the sensitivity of anthocyanins to the effect of the increase in temperature.

Conclusions

Although a decrease in the anthocyanin and polyphenol contents was expected with the increase in the drying temperature, this study was intended to quantify that decrease to establish the best conditions within the tested intervals. Thus, optimum spray drying conditions correspond to an air inlet temperature of 165 °C and 25% total solids. The optimized colorant presented 5.65% humidity, 25.2% hygroscopicity, poor fluidity, and high cohesiveness, with a dissolution time of 55 s, which allows its application in the food industry. The degradation of anthocyanins, adjusted to zero-order kinetics, was directly proportional to the increase in temperature and time. The values of the component a* decreased with increasing temperature and time. The natural food colorant obtained from *S. cumini* by spray drying presented potential technological and healthy properties due to its anthocyanins and polyphenols contents. Its application in food matrices such as fermented milk products should be evaluated, in which the pH will contribute to the stabilization of

Table 4 Kinetic parameters of anthocyanin loss and variation of component a* during accelerated storage of black cherry powder colorant

Indicator	Temperature (°C)	Model	R	R^2	R^2 adjusted	F	p	k*
Antho-cyanins loss	45	PA = 1,7414t-4,689	0,9776	0,9558	0,9495	151,3482	0,000,005	1,7414
	50	PA = 2,2122t + 5,4275	0,9886	0,9773	0,9744	343,7977	0,000,000	2,2122
	55	PA = 2,6763t + 19,015	0,966	0,9331	0,922	83,7291	0,000,096	2,6763
Compo-nent a* variation	45	ln a* = -0,0023t + 1,5164	0,9836	0,9674	0,962	177,9956	0,000,011	0,0023
	50	ln a* = -0,0078t + 1,5194	0,9934	0,9869	0,985	526,3325	0,000,000	0,0078
	55	ln a* = -0,0126t + 1,498	0,9768	0,9542	0,9485	166,6696	0,000,001	0,0126

Expressed as %.d^{-1} and d^{-1} for anthocyanin loss (zero order) and variation of the component a (first order), respectively anthocyanins and, therefore, the color of these products during storage.

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Declarations

Conflicts of interest The authors declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

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