

Influence of spray-drying on improving the quality of dried carob juice

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RESEARCH ARTICLE

Abstract

Spray-drying is a suitable method for drying carob juice, a popular drink, without a significant loss in beneficial nutritional properties or powder wetting behaviour. Maltodextrin (MD) was used as a wall material at different concentrations, namely 5, 10, 20 and 30% (MD dry solids/100 g feed mixture dry solids). Physical properties including moisture content, pH, titratable acidity, total soluble solids, and colour parameters were studied for the feed samples as well as the samples reconstituted after spray-drying. Moreover, the product yield, bulk density, and powder particle solubility were evaluated, as were their morphology and extent of nonenzymatic browning based on furfural content by using scanning electron microscopy (SEM) and solid-phase microextraction/gas chromatography-mass spectrometry (SPME/GC-MS). The increment in MD content increased the yield, solubility, and moisture, and decreased the bulk density of the spray-dried powder. Simultaneously, MD addition caused a decrease in pH values with an increase in the total soluble solids and titratable acidity of the reconstituted samples in comparison to the feed ones. As the MD to juice ratio increased, the lightness of the powders increased (L^*), while both colour parameters a^* and b^* decreased. Furfural extraction using SPME, followed by GC-MS analysis, showed a decrease in furfural concentration with the addition of MD. The presence of MD as the main carrier agent at a concentration of 20-30% enabled the formation of more homogeneous capsules with a better spherical shape and smoother surface, as observed under SEM. Therefore, MD can be successfully used at a 20-30% concentration to spray-dry carob juice and formulate an instant drink without any drawbacks.

Keywords: analytical methods, food quality, rheology, spray-drying, carob

1. Introduction

Carob fruit or carob pods belong to *Ceratonia siliqua* L. (carob tree), which is an evergreen tree cultivated mainly in the Mediterranean region and which recently spread to other areas, e.g. South America, Australia, India, and South Africa (Nasar-Abbas *et al.*, 2016). More than 315,000 tons of carob fruit are produced annually throughout the world (Boublenza *et al.*, 2017). People have been using carob for 4,000 years for its medicinal properties as an antiulcer, antidiarrheal, anti-inflammatory, phlegm-clearing, and stomach-strengthening agent, purgative, tonic, and more (Ben Hsouna *et al.*, 2012; Çakılcıoğlu and Türkoğlu, 2007; Jaradat, 2005; Kivçak *et al.*, 2002; Lev and Amar, 2002). Additionally, due to their nutritional importance, dried or roasted crushed carob pods can be used in powder form

to prepare beverages, syrups, and compotes, or as a cocoa substitute (Ayaz *et al.*, 2009).

Producing the popular carob beverage in instant dry powder form seems to be a promising technological approach for prolonging its shelf life, reducing its volume and weight and facilitating its handling (Akkaya *et al.*, 2012). Spray-drying is one of the most common industrial techniques for converting foods from liquid to powder form (Shrestha *et al.*, 2007). Tounsi *et al.* (2017) and Akkaya *et al.* (2012) have reported on the spray-drying of carob molasses; however, stickiness was the main problem encountered during the process, and this was associated with the higher sugar content (50-80%) of molasses. The addition of maltodextrin (MD) resulted in reducing the stickiness behaviour of spray-dried juices with respect to the air inlet temperature, which

affected the physicochemical properties of the formed powders (Quek *et al.*, 2007).

To the best of our knowledge, there have been no reports concerning the spray-drying of carob juice and the effect of the process on the quality of the instant product. Therefore, the aim of the present study was to evaluate the effects of the spray-drying process on the physicochemical properties of the final product.

2. Materials and methods

Materials and feed solution preparation

Roasted carob pods were purchased from the local market in Dokki, Giza, Egypt. The carob pods were first washed in water, sun-dried, manually fragmented, and finally ground to prepare carob juice (as an intermediate product) with water extraction as follows: carob pod grounds were soaked for 24 hours in water at room temperature, at a ratio of 1:4 kg/l. After extraction, the juice was first filtered through a filtering cloth. To spray-dry the carob juice, MD DE 15-20 (Yishui Dadi Corn Development Co., Shandong, China) was used as the drying agent. The feed solutions were prepared with juice to MD dry matter ratios of 95:5, 90:10, 80:20, and 70:30. The aqueous solutions were prepared by dispensing MD in carob juice at room temperature with constant stirring.

Methods

pH measurement

The pH values of the feed and reconstituted samples were measured in triplicate using a pH meter (Hanna HI 902 meter, Vöhringen, Germany). The concentration of each solution was 10% m/v.

Total soluble solids content

The total soluble solids of the feed and reconstituted samples was determined by measuring °Brix (0-32%) using a portable refractometer (ATAGO, Tokyo, Japan).

Titrateable acidity

The titrateable acidity was determined by the titration method (AOAC, 2005). The titrateable acidity (%) was expressed as citric acid equivalent.

Spray-drying process

A co-current Mini Spray Dryer B-290 (Büchi, Flawil, Switzerland) was used. The inlet and outlet temperatures were 160 and 80 °C (± 1.0 °C), respectively. The resulting powder was collected from both the drying chamber wall

and the cyclone. The drying process was conducted twice for each feed sample.

Powder characteristics

Yield

The spray-drying yield (%) was calculated by taking into consideration the total solid content of the feed sample with MD and the weight of the final dry powder (Saikia *et al.*, 2015).

Moisture content

Between 3 and 5 g of the powder were placed in a Petri dish, and the moisture was determined by heating at 105 °C until a constant weight was achieved. The percentage of powder moisture was calculated on a wet basis (AOAC, 2005). The arithmetic mean of three determinations was calculated \pm SD.

Solubility

Samples were mixed with distilled water (1:10 w/v), stirred for 1 hour at room temperature, and centrifuged at 1,500 rpm for 10 min. The supernatant was collected, dried, and weighed (Chau *et al.*, 2007).

Bulk density

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. The bulk density was calculated by dividing the mass of the powder by the volume it occupied in the cylinder. Duplicate samples were analysed and the mean reading was recorded (Karaaslan and Dalgıç, 2014).

Powder sample colour analysis

The extent of nonenzymatic browning (NEB) in the control (roasted carob pod) and spray-dried juice powders was measured by monitoring the formation of brown pigments, and expressed as the b^* value. The colour parameters (L^* , a^* , and b^*) were determined using a tristimulus colorimeter with the CIE-LAB colour space (Hunter Lab Scan XE, Reston, VA, USA) in reflection mode. The following parameters were measured: the L^* value – expressing lightness (in %); the a^* value – representing redness (positive) to greenness (negative); and the b^* value – indicating yellowness (positive) to blueness (negative). The colour difference was calculated as: $\Delta E = (\Delta a^{*2} + \Delta b^{*2} + \Delta L^{*2})^{0.5}$ (Farouk *et al.*, 2001).

Powder morphology using a scanning electron microscopy

The particle morphology of the spray-dried samples was analysed using a field emission scanning electron microscope (Quanta FEG 250, FEI, Prague, Czech Republic) at an accelerating voltage of 10 kV. The powder was previously gold sputtered by mounting on aluminium stubs with double-sided adhesive tape and coating with gold using an Edwards sputter coater S150A (Crawley, UK).

Furfural extraction using solid-phase microextraction

A 2 g sample of the spray-dried powders was introduced into a 20-ml solid-phase microextraction (SPME) vial. A 65 µm Carbowax™-Divinylbenzene (CW/DVB) coating fibre for a Manual Holder (Orange Label) from Supelco (Bellefonte, PA, USA) was used. The extraction was performed at 80 °C for 1 h. Then, the SPME fibre was inserted into the injector of a gas chromatography (GC) coupled to a mass spectrometry (MS) instrument. Desorption of the volatiles was performed at 220 °C for 2 min. The above method was reported and optimised by many authors (Farouk *et al.*, 2000).

Gas chromatography-mass spectrometry analysis

A Trace GC Ultra Chromatography system (Thermo Scientific, Waltham, MA, USA) equipped with an ISQ mass spectrometer (Thermo Scientific) was used for analysis. Volatile separation was performed on Supelcowax 10, film thickness 0.25 µm (Supelco). The oven temperature was maintained initially at 50 °C for 2 min, incremented from 50 to 220 °C at a rate of 2 °C/min, and finally kept isothermally at 220 °C for 30 min. The carrier gas was helium, with a constant flow rate of 1.0 ml/min. The injector temperature was 220 °C. The source and interface temperature was 250 °C. Mass Spectra in the electron impact model (EI) were obtained at 70 eV and a scan m/z range from 29 to 400 amu. Compound identification was performed by matching with the MS computer library (NIST 2005 version) and comparing with authentic compounds and published data. The relative percentages of carob powder volatiles were calculated from the GC peak areas. Kovat's index was calculated for each compound, using the retention times of a homologous series of C₆-C₂₆ n-alkanes and matching with literature (Adams, 2007).

Statistical analysis

The obtained data were subjected to a one-way analysis of variance (ANOVA). Duncan's test was used to detect significant difference at a level of 95%. Statistical analyses were made by SPSS software version 20.0. The data were expressed as mean ± SD.

3. Results and discussion

Physicochemical properties of the feed sample

The total soluble solids of the control carob juice sample was 9 °Brix, increasing significantly to 14-30 °Brix after the addition of 5-30% MD (Table 1). Acidity decreased significantly with the addition of MD, while the mixture pH values were not significantly affected and varied in the range of 5.48-5.53 (Table 1). The colour parameters of the ground carob pod powder are presented in Table 2 as a control sample.

Yield

The product yield varied in a range of 35.71-83.33% (Table 1). As seen, spray-drying carob juice with only 5% MD caused the materials to adhere to the wall chamber and the cyclone. The addition of MD changed the hygroscopic and thermoplastic characteristics of the powder. Therefore, the results indicate that yield increased significantly with the MD concentration, while the deposits on the dryer walls decreased. These findings are similar to the results of previous studies, which stated that carrier agents are necessary to produce powders suitable for consumption from juices with high sugar and acid content (Quek *et al.*, 2007).

Moisture content

The main physicochemical characteristics of spray-dried carob juice are presented in Table 1. To summarise, the current findings indicate significant differences ($P \leq 0.05$) in moisture as the MD concentration increased. The moisture values of the encapsulated juice varied from 3.7 to 5.63 wt%, which is considered to be within the same range that other investigations reported as acceptable for food industry applications (Kausadikar *et al.*, 2015). The recorded moisture contents in this study were found to be lower than in carob powders dried by microwaving (~7%) or roasting at 150 °C for 60 min (~9%), as reported by Tounsi *et al.* (2017) and Yousif and Alghazawi (2000), respectively. Therefore, spray-drying technology can be successfully used to prepare carob powders characterised by low water content and acid pH, which may contribute to long-term conservation and excellent protection against all bacterial and fungal alterations (Jridi *et al.*, 2015). As the MD concentration increased, the powder moisture increased, since an increase in the carrier agent concentration results in a decrease in time needed to form the particle shell, hindering water diffusion during the drying process (Tonon *et al.*, 2011).

Table 1. Physicochemical parameters of the feed and spray-dried and reconstituted carob juice samples.¹

Physico-chemical parameter	Feed samples					Reconstituted samples			
	Control (roasted carob pod powder)	Juice + 5% MD ²	Juice + 10% MD	Juice + 20% MD	Juice + 30% MD	Juice + 5% MD	Juice + 10% MD	Juice + 20% MD	Juice + 30% MD
pH	5.46±0.04 ^{b,c}	5.53±0.06 ^a	5.50±0.1 ^a	5.49±0.02 ^b	5.48±0.02 ^c	4.44±0.03 ^f	4.29±0.02 ^g	4.61±0.01 ^e	4.86±0.06 ^d
Titrateable acidity (%) (as citric acid)	0.194±0.02 ^f	0.185±0.01 ^g	0.174±0.07 ^h	0.165±0.01 ⁱ	0.152±0.06 ^j	3.192±0.04 ^a	2.629±0.07 ^b	1.232±0.01 ^c	0.942±0.02 ^d
Total soluble solids (°Brix)	9.00±0.05 ⁱ	14.00±0.02 ^h	16.50±0.07 ^g	23.00±0.01 ^e	30.00±0.02 ^c	19.0±0.08 ^f	25.00±0.03 ^d	31.00±0.01 ^b	43.00±0.13 ^a
						Powder samples			
Moisture content	–	–	–	–	–	3.70±0.02 ^d	4.52±0.08 ^c	4.72±0.01 ^b	5.63±0.13 ^a
Yield (%)	–	–	–	–	–	35.71±0.01 ^d	50.11±0.04 ^c	69.6±0.08 ^b	83.33±0.05 ^a
Solubility (%)	–	–	–	–	–	52.14±0.01 ^d	58.63±0.02 ^c	72.78±0.04 ^b	81.87±0.01 ^a
Bulk density (g/ml)	–	–	–	–	–	0.80±0.05 ^a	0.71±0.01 ^b	0.59±0.02 ^c	0.44±0.06 ^d

¹ Values represent averages ± standard deviations for triplicate experiments. Means with the same letter within the same row are not significantly different ($P>0.05$).

² MD = maltodextrin.

Solubility, total soluble solids, and bulk density

The solubility percentage of the samples ranged between 52.14 and 81.87% (Table 1). A higher MD content reduced browning, which simply consumes the sugar content of the carob (Table 2). Therefore, the high sugar content of the reconstituted samples with 20-30% MD may have contributed to their increased solubility (Papadakis *et al.*, 1998). Moreover, the smaller particle size and MD efficiency during the drying process, which were revealed by scanning electron microscopy (SEM) in this study, may be responsible for enhancing the solubility of the reconstituted samples

with 20-30% MD. The total soluble solids in °Brix showed a significant increase following the drying process, which may be due to the concentration of MD, followed by its hydrolysis (Mahendran, 2010).

An increase in the MD concentration led to a significant decrease in the bulk density of the reconstituted samples, from 0.80 to 0.44 mg/ml (Table 1), which is similar to the results obtained by Yousefi *et al.* (2011), Goula and Adamopoulos (2008), and Karaaslan and Dalgıç (2014) when pomegranate, tomato, and liquorice were dried using MD as a carrier. This phenomenon may be attributed to

Table 2. Colour parameters of carob juice powders.¹

Colour parameter	Control (roasted carob pod powder)	Carob juice powder			
		Juice + 5% MD ²	Juice + 10% MD	Juice + 20% MD	Juice + 30% MD
L*	44.75±0.01 ^e	68.42±0.02 ^d	74.28±0.01 ^c	81.53±0.00 ^b	94.85±0.07 ^a
a*	12.69±0.04 ^a	5.08±0.02 ^b	3.49±0.01 ^c	3.76±0.01 ^d	0.06±0.01 ^e
b*	21.75±0.02 ^d	30.25±0.03 ^a	28.00±0.03 ^b	24.32±0.00 ^c	18.23±0.01 ^e
ΔE	–	26.27±0.06 ^d	31.55±0.02 ^c	37.93±0.03 ^b	51.78±0.04 ^a

¹ Values represent averages ± standard deviations for triplicate experiments. Means with the same letter within the same row are not significantly different ($P>0.05$).

² MD = maltodextrin.

the fact that MD addition minimizes the stickiness of thermoplastic particles with an increase in the volume of air trapped inside the particles, which causes a decrease in the apparent density of the particles (Goula and Adamopoulos, 2010).

pH and acidity of reconstituted samples

A significant decrease in pH values was observed for the reconstituted samples produced by spray-drying in comparison to the mixture of fresh juice samples and MD (Table 1). The pH of the fresh juice/MD samples varied between 5.46 and 5.53, while the pH of the reconstituted samples ranged between 4.29 and 4.86; therefore, the reconstituted juices are classified as low-acid food. In agreement with the above results, a dramatic increase in titratable acidity was seen (Table 1) that may be due to the MD concentration being accompanied by release of sugars and other constituents during drying (Chopda and Barrett, 2001; Saikia *et al.*, 2015).

A perusal of the literature revealed that spray-drying various types of juices caused significant variation in the pH values of the reconstituted juice. The pH value of pineapple fruit juice increased with the addition of MD; the same trend was also observed in reconstituted pineapple juice, as well as in liquorice extract and guava juice (Hashib *et al.*, 2015; Karaaslan and Dalgıç, 2014; Mahendran, 2010). Saikia *et al.* (2015) showed a dramatic decrease in the pH value of carambola reconstituted juice (pH 1.98) in comparison to the feed sample (pH 2.77), while the changes in pineapple, watermelon, and mandarin were not significant. However, there have been no reports concerning the reasons for the variation in pH due to spray-drying. The wall materials used in encapsulation may also affect the pH of the final product, as reported by Khuenpet *et al.* (2016): they found that using MD as a drying aid had a significant ($P < 0.05$) effect on pH, while the samples encapsulated with Arabic gum were not significantly ($P > 0.05$) different from the fresh juice. Finally, they stated that further study was required for a better understanding of this phenomenon. In the present study, the pH decrease of the spray-dried carob juice was accompanied by NEB or caramelisation, as shown by the colour parameters below (Table 2), which is in agreement with Van Boekel (2001) who showed a substantial reduction in pH at higher temperatures due to increased water dissociation.

Colour analysis of powder samples

The colour measurement results for the spray-dried powders are shown in Table 2. In comparison to the control sample, the lightness of the powders increased significantly (increasing L^*) as the MD to juice ratio increased, which was a result of the lower juice content. The yellowness level, represented by b^* , was significantly higher than for the

control sample, while a decrease in the redness level, defined by a^* , was observed (Table 2). A significant decrease in the same parameters was observed for higher concentrations of MD (20 and 30%), which seems to inhibit the Maillard reaction or NEB due to a more effective encapsulation. As a consequence, significant colour differences (ΔE) could be observed due to the respective changes in the colour parameters (Table 2).

The changes in a^* and b^* correspond to the initial increase in redness and yellowness, respectively, which occurs in the beginning of the NEB reaction (or Maillard reaction), while products with more intense green and blue colour notes are formed only later, as a result of secondary reactions. These cannot occur over the course of a very short encapsulation time and at relatively low temperatures (120–160 °C) (Farouk *et al.*, 2001). In this respect, the spray-dried juice that contained only 5% MD was characterised by a dark brown colour ($L^* = 68.42$, $a^* = 5.08$, $b^* = 30.25$), which is in agreement with the results reported by Boublenza *et al.* (2017) for roasted carob pod powder. By contrast, the powder sample containing 30% MD was characterised by a light brown colour ($L^* = 94.85$, $a^* = 0.06$, $b^* = 18.23$). The effect of the MD amount on the encapsulation of carob molasses as well as their physical properties was studied by Akkaya *et al.* (2012). They found that an increase in the amount of MD increased the powder lightness and decreased both a^* and b^* , and vice versa, which is in agreement with the results of the present study.

Nonenzymatic browning and furfural indicator

The NEB, or Maillard, reaction is responsible for the principal changes in the physical, chemical, and sensory properties of carob during thermal treatment, e.g. roasting or spray-drying. Boublenza *et al.* (2017) showed a gradual degradation in the carob sugar content with heating and roasting time. Sugars undergo breakdown and dehydration based on the NEB or Maillard reaction, which is divided into three main stages: the early, intermediate, and final stage. Taking into account the short encapsulation time and lower temperature, only the early and intermediate stages will be discussed here. These stages are relevant for aroma formation, e.g. furfural, which is presented in this study as a marker for browning due to its easy extraction and analysis using SPME-GC/MS. In the early stage, the Schiff base formed through sugar-amine condensation undergoes acid-catalysed rearrangement via an eneaminol to produce the Amadori compound, which is relatively stable in its cyclic furanose form and can be isolated (Teranishi *et al.*, 1998). In the intermediate stage and at low pH, the 1,2-enolisation of the Amadori compound is favoured, followed by loss of water, and hydrolysis of the imine releases the amino acid (intact) and the 3-deoxyhexosulose. The major breakdown products of the latter are 5-hydroxymethylfurfural, which results from dehydration and cyclisation, formic acid, and

2-furfural, which forms by hydrolytic α -dicarbonyl cleavage, in addition to methylglyoxal and glyceraldehyde, which are generated via retroaldolisation (Brands and Van Boekel, 2001).

In addition to the 1,2-enolisation pathway for Amadori compounds derived from sugars, furfural is also produced through the NEB of ascorbic acid, a vitamin reported to be present at high levels in carob (Youssef *et al.*, 2013).

5-hydroxymethylfurfural was evaluated as a quality parameter for carob molasses by Tetik *et al.* (2010) using HPLC, while Farag and El-Kersh (2017) identified furfural at 0.34% among 31 volatile constituents of roasted carob pods using headspace (HD) SPME analysed via a quadruple mass spectrometer. The previous finding is in agreement with the present study, where 0.399% furfural was detected in the control sample. Figure 1 illustrates the GC-MS

chromatograms for the volatiles of the spray-dried samples, with a focus on furfural, which was extracted using SPME.

The furfural content was found to gradually decrease from 1.247 to 0.401% as the MD amount increased from 5 to 30%, which revealed the encapsulation efficiency of MD and the reduction in NEB, and agreed with the colour parameter results (Table 2, Figure 2).

The detection of higher furfural concentrations was expected in the present work as compared to the roasted carob pods studied by Farag and El-Kersh (2017), due to the NEB reaction accompanying spray-drying. Furfural is often used as a browning indicator for many food products, including citrus juice, fruits, infant formula, etc. (Guerra-Hernandez *et al.*, 2002; Shinoda *et al.*, 2005; Yamada *et al.*, 2009). Therefore, it was looked at here as a scaling indicator for carob browning during spray-drying, using

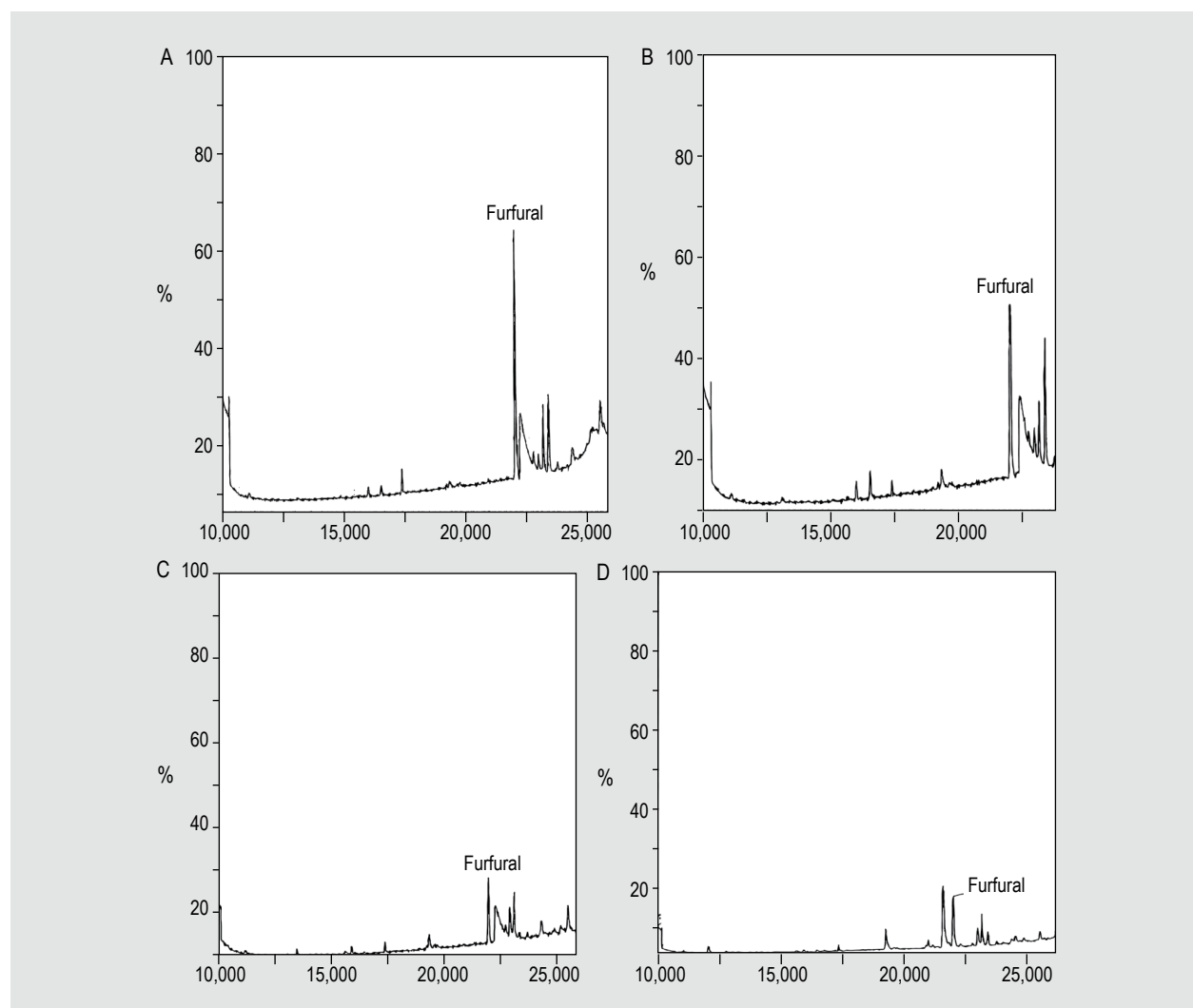


Figure 1. Gas chromatography-mass spectrometry chromatograms of furfural extracted from powder samples with (A) 5% maltodextrin (MD); (B) 10% MD; (C) 20% MD; and (D) 30% MD as wall material.

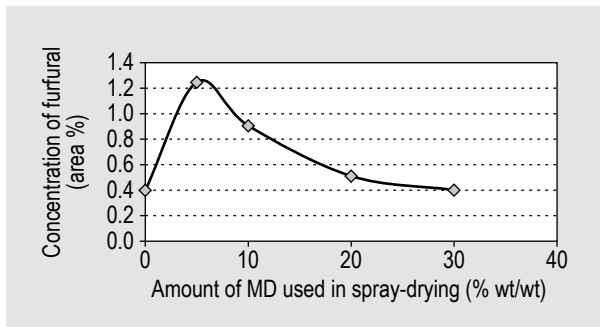


Figure 2. Effect of maltodextrin (MD) addition on furfural formation as non-enzymatic browning indicator.

SPME as a fast and reliable extraction technique with no known downsides.

Surface morphology of powder samples

Based on their sugar content, spray-dried products can be classified as non-sticky and sticky. Fruit and vegetable juice, honey, and lactose powders are categorised as sticky products due to their high content in sugars, namely glucose, fructose, and sucrose, and organic acids, i.e. citric, ascorbic, and tartaric acid (Adhikari *et al.*, 2004). It is well-known that biopolymers such as MD are successfully used as wall materials in order to achieve a

qualified product and overcome stickiness during spray-drying (Tan *et al.*, 2011). In the present study, stickiness was observed clearly in spray-dried carob juice containing 5% MD, and it disappeared completely in the spray-dried samples containing 20 and 30% MD. Such stickiness is reflected in the surface morphology of the dried powders, with a fused surface, clumped particles, and complex link bridges being visible in the sticky samples containing 5 and 10% MD (Figure 3A and 3B). The same phenomena were observed by Yousefi *et al.* (2011) during the encapsulation of pomegranate juice using different carriers, including MD, in lower amounts, not exceeding 12%. SEM micrographs (Figure 3C and 3D) show that using MD as a carrier agent at a level of 20 and 30% enabled the formation of more homogeneous capsules with a better spherical shape and smoother surface. The formation of some indentations on the surface of particles obtained by spray-drying is usually attributed to particle shrinkage due to drastic loss of moisture followed by cooling (Saéñz *et al.*, 2009). Different SEM patterns were reported by Saikia *et al.* (2015) for various encapsulated juices using 20% MD: whereas both small and large spherical particles were observed in spray-dried carambola juice; large, globular, and ellipsoidal particles were found in watermelon powder; and finally, fused particles with link bridges were seen in pineapple and Khasi mandarin powders.

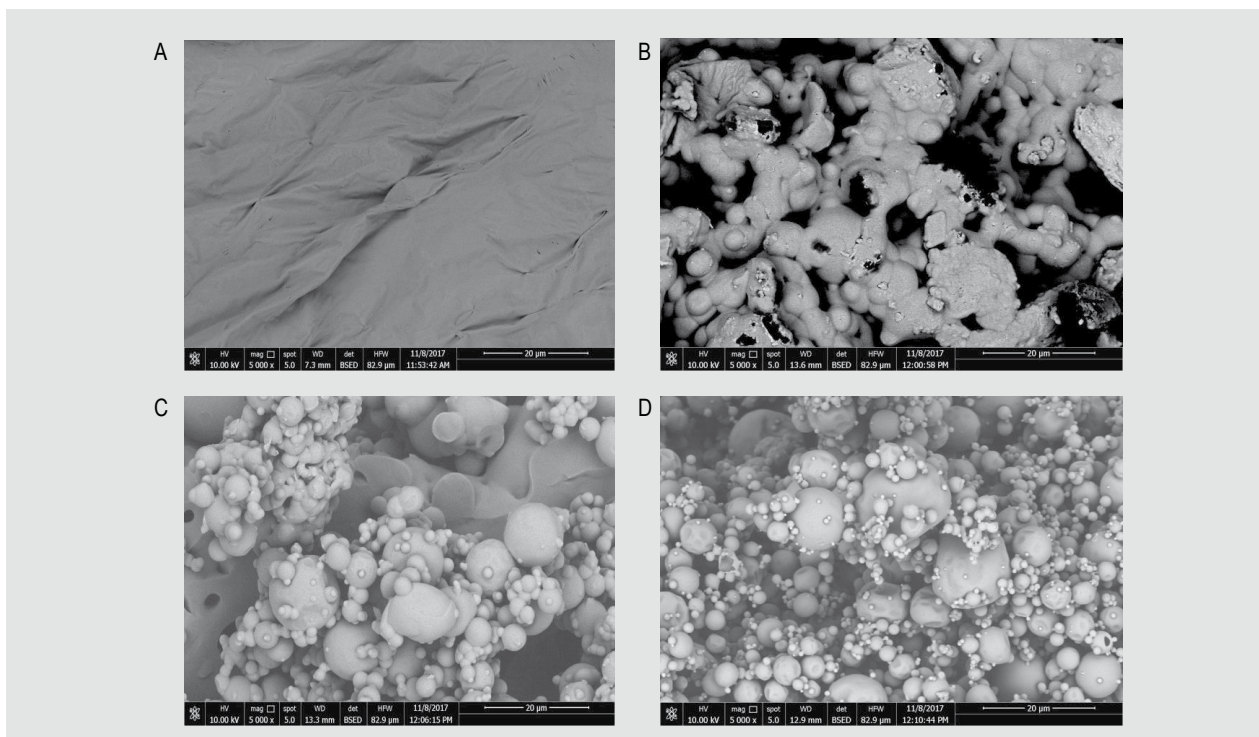


Figure 3. Scanning electron microscope of spray-dried samples produced with (A) 5% maltodextrin (MD); (B) 10% MD; (C) 20% MD; and (D) 30% MD as wall material.

4. Conclusions

The spray-drying process and MD concentration had an effect on the physicochemical properties, morphology, and browning characteristics of the end product, and therefore on the amount of compounds generated through the Maillard reaction, e.g. furfural. The product yield, total soluble solids, and solubility increased while the bulk density decreased with the addition of MD, which changed the hygroscopic and thermoplastic characteristics of the powder and minimized browning. Higher MD concentrations increased the moisture content, which remained within the acceptable range for the food industry. A significant decrease in pH values was observed in spray-dried samples, which was usually accompanied by a browning reaction at a higher temperature with an increase in water dissociation. According to the different CIE-LAB characteristics, an increase in MD reduced browning by lowering furfural formation. SEM showed that the presence of more MD enabled the formation of homogeneous capsules with a better spherical shape and smoother surface. Drying process using 20-30% MD can be used in order to successfully prepare a spray-dried carob juice product with acceptable physicochemical properties and without browning or stickiness problems.

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