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Physico-Chemical Characterization of Encapsulated Fennel Essential Oil under the Influence of Spray-Drying Conditions

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Abstract: In this study, fennel essential oil (EO) was spray-dried, varying the wall material type (two-component blends of maltodextrin (MD), β-cyclodextrin (β-CD) and gum arabic (GA)), the wall material ratio (1:1, 1:3 and 3:1) and the drying temperature (120, 160 and 200 °C). A total of 27 powders were analyzed for their moisture content, solubility, hygroscopicity, bulk density and particle size, while powder recovery and oil retention were determined in terms of encapsulation efficiency. The morphology and chemical composition of the powder obtained under optimal conditions were additionally analyzed by scanning electron microscopy and gas chromatography-mass spectrometry. The results showed that all of the powders had generally good properties, exhibiting a low moisture content, high powder recovery and high oil retention. A 1:3 MD:GA mixture and a drying temperature of 200 °C were found to be optimal for the spray-drying of fennel EO, producing a powder with a low moisture content (3.25%) and high solubility (56.10%), while achieving a high powder recovery (72.66%) and oil retention (72.11%). The chemical profiles of the initial and encapsulated fennel EO showed quantitative differences without qualitative changes, with an average 24.2% decrease in the volatiles in the encapsulated EO. Finally, spray-drying proved to be a successful tool for the stabilization of fennel EO, at the same time expanding the possibilities for its further use.

Keywords: *Foeniculum vulgare* Mill.; essential oil; encapsulation; powder; wall material; oil retention; maltodextrin; β-cyclodextrin; gum arabic

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1. Introduction

Fennel (*Foeniculum vulgare* Mill.) seeds are known for their richness in essential oil (EO), which can even be up to 5% [1]. Fennel EO is described as a clear, pale-yellow liquid characterized by a distinctive, strong, anise-like flavor for which various volatile compounds, mainly anethole, are responsible [1]. In addition to its notably perceptible sensory properties, numerous studies have demonstrated and confirmed its diverse biological activities including antimicrobial, antiviral, anti-inflammatory, antiallergic, antimutagenic, hepatoprotective, etc. [2]. Precisely because of its flavoring and therapeutic properties, fennel EO is often used as a flavoring agent in the food industry, as well as for cosmetic and pharmaceutical purposes [3,4]. However, like other essential oils, fennel EO reacts sensitively to external factors such as oxygen, light, moisture and heat due to its volatile chemical structure. Therefore, the essential oils should be stored in tightly closed, dark-colored glass bottles in a cool dark place and protected from direct light when on the market. However, against this background, it is a challenge to extend its shelf life and enable longer use, especially as fennel EO is widely used. One of the promising tools to prolong the stability of essential oils is certainly encapsulation by spray-drying.

Spray-drying is a well-known and broadly applied encapsulation technique, especially in the food industry [5]. It enables the conversion of a liquid form into a dry form of powder by drying in a stream of hot gas, encapsulating the active ingredient (core material) in a protective layer of wall material (carrier) and thus forming micro- or macroparticles. This not only protects the active substance from the negative effects of environmental conditions, but also allows its alternative and much wider use. Spray-drying has therefore been extensively used in the food, cosmetic and pharmaceutical areas, particularly due to relatively inexpensive and accessible equipment. One should mention that notably great scientific attention has been paid to the encapsulation of natural bioactive compounds. Numerous studies have documented a successful application of spray-drying in the encapsulation of polyphenols, vitamins, carotenoids, chlorophylls, phytosterols and fatty acids [6–10], whereby the predominant focus was set on polyphenols. The topic of encapsulation of essential oils by spray-drying has also been covered by scientific data, although to a much lesser extent. Existing studies on the spray-drying of essential oils show examples of spray-drying of the EO of rosemary [11,12], thyme [13], oregano [14], cinnamon [15], spearmint [16], lemongrass [17], lavender [18] and coriander [19], but to the best of authors' knowledge there are no data on the spray-drying of fennel EO.

Spray-drying can be considered a convenient drying method for delicate essential oils, since it has been specifically utilized for heat-sensitive substances. Although EO components are volatile and susceptible to heat, and the temperatures used in spray-drying (>100 °C) could have negative effects, the loss of volatiles during spray-drying is reduced by the short exposure of the core material to heat, as the rapid evaporation of the liquid from the surface of microparticles during the solidification of the wall material coating keeps the temperature of the core material below 100 °C [20]. The process itself involves following these main steps: preparation of the emulsion; homogenization; atomization of the emulsion; and dehydration of the atomized particles [21]. The first step refers to the dissolution of the appropriate wall material in a suitable solvent (usually water), whereby it should be noted that the hydrophobic properties of EO require the use of hydrophilic wall material. In the second step, the core material is mixed with the wall material suspension to form a stable emulsion, which is often achieved by adding emulsifiers and/or high-speed stirring, given the water affinity of the EO components. The resulting solution is then atomized and dried in a stream of hot gas (usually air, rarely nitrogen) in the drying chamber of a spray-dryer, finally producing a powder [20,22] consisting of spherical particles [23]. However, in order to obtain a satisfactory yield of encapsulated EO with the desired physico-chemical characteristics, i.e., a powder consisting of particles of uniform size with low moisture content and hygroscopicity, high solubility and bulk density, and with minimal or no change in chemical composition compared to the initial raw material, it is essential to define the optimal process settings that ensure all the above properties. In view of this, the choice of wall material plays an unavoidable role.

When selecting the wall material, its physical and chemical properties should be considered, as any decomposition or interaction of the final product with the wall material is unacceptable [24,25]. Moreover, the wall material should be nontoxic, inexpensive, biodegradable and highly soluble with good emulsifying, drying, film-forming and protective properties [23]. For the purposes of flavors encapsulation, a wide range of wall materials are usually used: polysaccharides (starches, maltodextrins, gum arabic, cyclodextrins and corn syrups), lipids (mono- and diglycerides), proteins (casein, milk serum and gelatin) and new emerging biopolymers [26]. Of the wall materials listed, maltodextrin (MD) is most commonly applied for spray-drying. It is a polysaccharide formed of multiple D-glucose units linked by α-1,4-glycosidic bonds obtained by the partial hydrolysis of corn starch, described with dextrose equivalent (DE), which marks the hydrolysis level and is directly related to the degree of protection of the core material against the penetration of oxygen. MD is characterized by a relatively low cost, a neutral aroma and taste, low viscosity and hygroscopicity, high water solubility and good protective effect against the oxidation of the encapsulated EO. Due to its poor emulsifying properties and the retention

of volatiles, it is usually combined with other wall materials [13,27]. Cyclodextrins are polysaccharides obtained from the breakdown of starch by enzymes and occur in three different forms: α-, β- and γ-cyclodextrin, with particular attention during encapsulation being paid to β-cyclodextrin (β-CD). A β-CD molecule consists of seven glucopyranose units in the form of a truncated cone, whose outer surface is hydrophilic due to many hydroxyl groups, and the inner cavity is hydrophobic. This structure enables the solubility of β-CD in water, while the hydrophobic substances of the core material are enclosed in an inner cavity [20,28]. Another frequently used wall material in spray-drying is gum arabic (GA), which is obtained from the *Acacia* tree. It is a polymer consisting of D-glucuronic acid, L-rhamnose, D-galactose and L-arabinose, with almost 2% protein [29,30]. It acts as an emulsifier, stabilizer, flavoring agent, humectant, thickening agent, surface refining agent and to delay sugar crystallization, and it ensures a high retention of volatiles during spray-drying [29]. However, its use also often implies a combination with other wall materials, as its disadvantages are reflected in its high cost, limited availability, contamination and inability to prevent oxidation [23,31]. To eliminate the drawbacks of wall material individual use, and to maximize the encapsulation efficiency and positive properties of the encapsulated EO, a two- or multi-component mixture of wall material in varying proportions is often used [32]. In order to promote the best properties of the wall materials when they are combined, it is certainly necessary to determine their optimal ratio to each other.

Another decisive parameter for effective spray-drying is definitely the drying temperature or the inlet temperature. The temperatures used for spray-drying usually exceed 100 °C which results in instant evaporation. Thereby, a sufficiently high temperature allows a faster drying process reducing the surface tension and viscosity, leading to the rapid development of a semi-permeable crust on the outer layer of the droplets, which consequently entraps the core material inside the particles. Determining the appropriate drying temperature is extremely important, as the application of a low temperature leads to slower evaporation, so that the particles have a high membrane density, high moisture content, low flowability and easy agglomeration, which in turn leads to a low process yield as the particles adhere to the inner wall of the drying chamber. On the other hand, extreme evaporation occurs at excessively high temperatures, which can cause cracks in the membrane, leading to leakage of the core material and thus to its decomposition or exhaustion [20,22,33].

As mentioned above, there are no scientific data on the encapsulation of fennel EO by spray-drying or the physico-chemical properties of fennel EO powders obtained by this encapsulation method. Therefore, the goal of this study was to examine the influence of the wall material type (two-component mixtures of MD, β-CD and GA), the wall material ratio (1:1, 1:3 and 3:1) and the drying temperature (120, 160 and 200 °C) during spray-drying on the process yield and efficiency, as well as the physico-chemical properties of the obtained fennel EO powders.

2. Materials and Methods

2.1. Chemicals

Purified water was of Milli-Q quality (Millipore, Bedford, MA, USA) and *n*-hexane 95% was procured from Fisher Scientific (Loughborough, UK). Sodium chloride and anhydrous sodium sulfate were obtained from Lach-Ner Ltd. (Neratovice, Czech Republic). Commercial standards of α-pinene, camphene, β-pinene, α-phellandrene, 3-carene, *p*-cymene, γ-terpinene, eucalyptol, L-fenchone, camphor, fenchyl acetate, carvone *p*-anisaldehyde, *trans*-anethole and alkane standard solution C_7–C_{30} were purchased from Sigma Aldrich (St. Louis, MO, USA); myrcene, linalool and α-terpineol from Merck (Darmstadt, Germany); D-limonene and nerol from Fluka® Analytical (Munich, Germany); and α-terpinene and estragole from Dr. Ehrenstorfer GmbH (Augsburg, Germany).

2.2. Material

Fennel (*F. vulgare* Mill.) EO procured from Ireks Aroma Ltd. (Jastrebarsko, Croatia) was used as the core material. Fennel EO was obtained from the seeds of bitter fennel by hydrodistillation and had the following properties: refractive index at 20 °C of 1.530–1.550, density at 20 °C of 0.960–0.980 g/cm^3 and flash point of 63 °C. The wall materials used were MD DE 4–7 (Biosynth, Bratislava, Slovakia), GA (Alfa Aesar, Ward Hill, MA, USA) and β-CD (Acros Organics, Geel, Belgium). Tween® 20 from AppliChem GmbH (Darmstadt, Germany) was used as an emulsifier.

2.3. Preparation of Emulsions

Wall materials in a mixed form of MD:β-CD, MD:GA and β-CD:GA at a specified ratio (1:1, 1:3 and 3:1, *w*/*w*) were dissolved in 200 mL of distilled water under constant stirring at 600 rpm and 50 °C (RT 5, IKA-Werke, Staufen im Breisgau, Germany). Prepared mixtures were kept overnight to ensure the full saturation of the polymer molecules. Then, fennel EO and Tween® 20 were progressively added to the wall material solution under stirring at 10 000 rpm for 5 min using Ultra-Turrax IKA T25 D (Staufen, Germany) to form an emulsion. Emulsions were prepared with 10 g of EO, 5% addition of Tween® 20 with respect to the EO weight and an appropriate amount of wall material to fulfil the mass ratio of EO to wall material of 1:4 (*w*/*w*) in the total emulsion volume. Freshly prepared emulsions were used as feed solutions in the spray-drying process.

2.4. Preparation of Encapsulated Essential Oil by Spray-Drying

The emulsions were subjected to drying in a laboratory-scale Mini Spray Dryer B-290 (Büchi, Flawil, Switzerland) under the following constant parameters: compressed air flow rate 28 m^3/h, feed flow rate by a peristaltic pump 150 mL/h and nozzle cleaner set at Level 4. The inlet air temperatures (120, 160 and 200 °C) were varied according to the experimental design. The obtained powders were collected and stored in airtight plastic containers in a desiccator at 20 °C for further analysis. All powders were produced in duplicate.

2.5. Characterization of the Encapsulated Fennel Essential Oil

2.5.1. Moisture Content

The moisture content of the obtained EO powders was determined by drying at 105 °C until constant weight [34]. The percentage was calculated as a mass ratio before and after drying.

2.5.2. Solubility

The solubility of the EO powders was determined according to the method proposed by Anderson et al. [35] with some modifications. Briefly, EO powder (1 g) was stirred in distilled water (10 mL) for 1 min using vortexer, thermostated in a water bath (B-490, Büchi, Flawil, Switzerland) at 37 °C/30 min and afterwards centrifuged (Rotofix 32, Hettich, Kirchlengern, Germany) at 5500 rpm/20 min. The resulting supernatant was oven-dried at 105 °C until constant weight. Solubility was calculated by the weight difference according to the following Equation (1):

$$Solubility\ (\%) = \frac{m_s}{m_p} \times 100 \quad (1)$$

where m_s is the mass of EO powder obtained by drying the supernatant to a constant weight (g) and m_p is the mass of EO powder taken for analysis (g).

2.5.3. Hygroscopicity

For hygroscopicity, the method by Tonon et al. [36] was used. An amount of 1 g of each powder sample was placed in an open Petri dish in an airtight vessel containing saturated

NaCl solution (75.29% humidity) at 25 °C. After 1 week, the samples were weighed, and hygroscopicity was determined as the weight in g of the adsorbed moisture per 100 g of dry solids (g/100 g) according to the following Equation (2):

$$Hygroscopicity\ (\text{g}/100\ \text{g}) = \frac{m_7 - m_0}{m_0} \times 100 \quad (2)$$

where m_7 is the mass of the weighed microcapsules after 7 days (g) and m_0 is the initial mass of the microcapsules (g).

2.5.4. Bulk Density

The bulk density of EO powders was evaluated by the method of Beristain et al. [37]. An amount of 2 g of powder was put into 10 mL graduated cylinder and held on a vortexer for 1 min to distribute the powder particles uniformly. The cylinder was then placed on a flat surface and the final volume of the powder was recorded. The bulk density was calculated as the ratio between powder mass and the volume occupied in the cylinder (g/mL).

2.5.5. Particle Size Distribution

The particle size distribution of the EO powders was measured by laser diffraction by the Malvern Mastersizer particle size analyzer assembled with the dry dispersion unit Scirocco 2000 (Malvern Instruments, Worcestershire, UK). An amount of approximately 3 g of the powder sample was added to the dry dispersion unit, and measurements were taken at a pressure of 1.5 bar and a flow rate of 100%. Each measurement was carried out in duplicate and the diameter of the 50th percentile [d (50), µm] was calculated by the software Mastersizer 2000 v. 5.60 (parameters: refractive index of 1.5, absorption of 0.1 and obscuration of 3%). A value of d (50) was considered to assess the particle size distribution of the EO powders.

2.5.6. Powder Recovery

The powder recovery, or spray-drying process yield, was calculated as the ratio between the dry matter content of the powder obtained after spray-drying and the dry matter content of the initial emulsion according to the following Equation (3) [38]:

$$Powder\ recovery\ (\%) = \frac{m_p}{m_{eo} + m_{em} + m_c} \times 100 \quad (3)$$

where m_p is the mass of the produced EO powder (g), while m_{eo} is the mass of the EO (g), m_{em} is the mass of the emulsifier (g) and m_c is the mass of the carrier (g) in the initial emulsion.

2.5.7. Oil Retention

Oil retention was determined according to the modified method of Marques et al. [13]. Briefly, 10 g of EO powder and 200 mL of distilled water were mixed and subjected to hydrodistillation for 120 min using a Clevenger-type apparatus. The obtained EO was dried over anhydrous sodium sulfate.

Oil retention was defined as the ratio between the mass of EO present in the powder and the initial mass of oil taken for the drying process, and it was calculated according to the following Equation (4):

$$Oil\ retention\ (\%) = \frac{total\ oil\ extracted\ (g)}{initial\ oil\ load\ (g)} \times 100 \quad (4)$$

2.5.8. Morphology of Encapsulated Fennel Essential Oil by Scanning Electron Microscopy (SEM) Analysis

The morphological characteristics of the encapsulated fennel EO optimal formulation were studied using a high-resolution field emission scanning electron microscope (SEM) JSM-7000F (Jeol, Tokyo, Japan). The samples were applied in a thin layer on a conductive carbon tape on the SEM sample holder. The images were taken with an accelerating voltage of 5.0 kV at a standard distance between objective and sample (WD = 10 mm). The photomicrographs were taken at 500× and 1000× magnification, and a secondary electron detector was used to create the micrograph/image.

2.5.9. Gas Chromatography—Mass Spectrometry Analysis

To evaluate the composition of the fennel EO before spray-drying and one hydrodistilled from the encapsulated EO optimal formulation, gas chromatography—mass spectrometry (GC-MS) analysis was performed by the method of Marčac et al. [1]. For the analysis, an Agilent Technologies 6890N Network GC System equipped with an Agilent 5973 *inert* mass selective detector (Agilent Technologies, Santa Clara, CA, USA) and a capillary column Agilent HP-5MS [(5%-phenyl)-methylpolysiloxane; 30 m × 0.25 mm × 0.25 μm] were used. Samples diluted (1:99) in a mixture of *n*-hexane and internal standard (nerol, 1.0518 mg/mL) were automatically injected (1.0 μL, Agilent 7683B autosampler injector) in the split mode at a ratio of 1:100 and injection temperature of 250 °C. Helium was used as the carrier gas at a constant flow rate of 1 mL/min. The program of the oven temperature was set from 60 to 145 °C at the rate of 3 °C/min and 145 to 250 °C at the rate of 30 °C/min, being held at the final temperature for 3 min. The transfer line, MS source and quadrupole temperatures were kept at 280, 230 and 150 °C, respectively. The detector ionization energy was set at 70 eV. The mass spectra (m/z) were recorded in a range of 30–550 at 1 scan/s, and quantification was performed in single ion monitoring (SIM) mode. The same conditions were applied for the analysis of the alkane solution, and retention indices (RI) were calculated according to Bianchi et al. [39].

EO components were identified by comparison of their retention times, RI and m/z with authentic standards and by comparing with the m/z available in the NIST library (ChemStation Data Analysis). Volatiles were quantified using calibration curves of the α-pinene, camphene, β-pinene, myrcene, α-phellandrene, α-terpinene, *p*-cymene, D-limonene, eucalyptol, γ-terpinene, L-fenchone, linalool, camphor, α-terpineol, estragole, fenchyl acetate, carvone, *p*-anisaldehyde and *trans*-anethole. Sabinene and *cis*-sabinene hydrate were identified according to their m/z, RI and comparison with the literature data, while the 3-carene calibration curve was used for their quantification. The results are expressed in mg/mL of the EO as the mean ± standard deviation (SD).

2.6. Experimental Design and Statistical Analysis

Statistica ver. 10.0 software (Statsoft Inc., Tulsa, OK, USA) was used for experimental design and statistical analysis. All analyses were carried out in duplicate. A three-level full factorial experimental design consisting of 27 trials was used to evaluate the effects of three independent variables, namely the wall material type (MD:β-CD, MD:GA and β-CD:GA), the wall material ratio (1:1, 1:3 and 3:1) and the drying temperature (120, 160 and 200 °C), on the moisture content, solubility, hygroscopicity, bulk density, particle size, powder recovery and oil retention (dependent variables). The normality of the data and homoscedasticity of the residuals were tested by Shapiro–Wilk's test and Levene's test, respectively. Normally distributed and homogeneous data were analyzed using a multifactorial analysis of variance (ANOVA), whereas marginal means between groups were compared using Tukey's HSD multiple comparison test. Nonparametric data were analyzed with the Kruskal–Wallis test. The results of statistical analysis are presented as the mean ± standard error (SE). Principal component analysis (PCA) was performed for the fennel EO powders' properties using the principal components (PC) with an eigenvalue > 1, including variables with communalities ≥ 0.5. Significant differences in the chemical

composition of the initial EO before spray-drying and the encapsulated EO obtained under optimal spray-drying conditions were tested using one-way ANOVA and a post-hoc Tukey's HSD test, and the results are presented as the mean ± SD. The significance level $p \leq 0.05$ was assigned for all tests.

3. Results and Discussion

In order to produce encapsulated fennel EO in a powder form, thereby preserving its quality and promoting its potential further use, this study aimed to evaluate the influence of the spray-drying conditions on the process yield and the physico-chemical properties of the obtained EO powders. Therefore, fennel EO was spray-dried in combination with different blends of wall material (MD:β-CD, MD:GA and β-CD:GA) mixed in different ratios (1:1, 1:3 and 3:1) at three drying temperatures (120, 160 and 200 °C), and the moisture content, solubility, hygroscopicity, bulk density and particle size of the obtained powders, as well as the powder recovery and oil retention, were determined. The results obtained are shown in Table 1, while Table 2 provides an overview of the influence of the spray-drying conditions tested on the powder properties listed. The distribution of the EO powders according to their properties analyzed by PCA is shown in Figure 1. Moreover, the morphology of the EO powder obtained at the optimal spray-drying conditions and its chemical composition, which was additionally compared with the initial EO used for spray-drying, were also examined. The results reflecting these analyses are summarized in Figures 2 and 3 and Table 3, respectively.

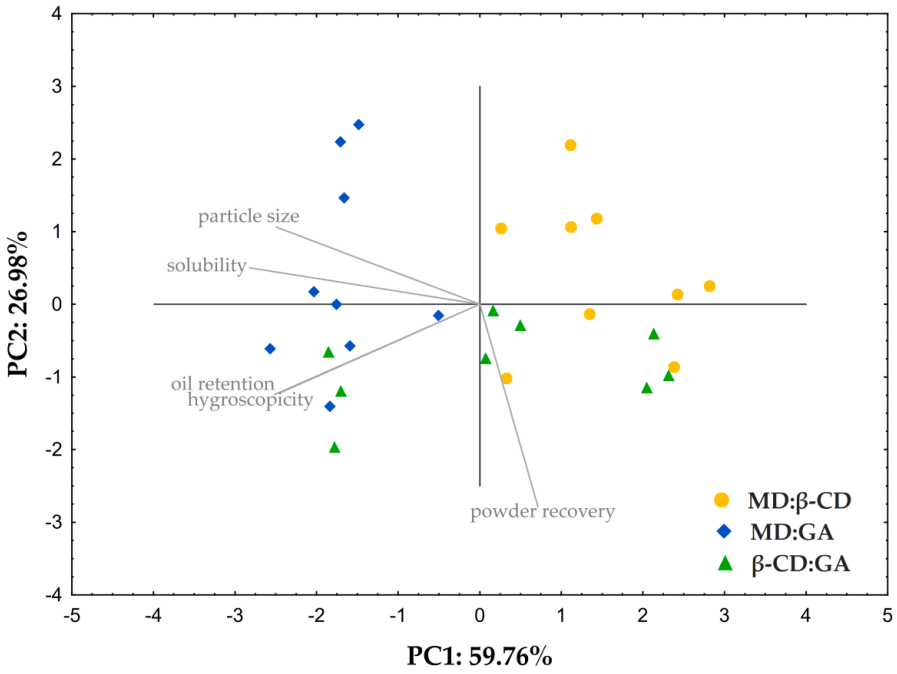


Figure 1. Distribution of encapsulated fennel essential oil in a two-dimensional coordinate system defined by the first two principal components (PC1 and PC2) in relation to the type of wall material.

Table 1. Physical properties of encapsulated fennel essential oil obtained under different spray-drying conditions.

Assay No.	Wall Material Type	Wall Material Ratio	Drying Temperature (°C)	Moisture Content (%)	Solubility (%)	Hygroscopicity (g/100 g)	Bulk Density (g/mL)	Particle Size [d (50), µm]	Powder Recovery (%)	Oil Retention (%)
1	MD:β-CD	1:1	120	4.86 ± 0.16	41.44 ± 0.85	8.84 ± 0.05	0.40 ± 0.03	8.59 ± 0.03	65.95 ± 0.51	32.68 ± 0.62
2			160	3.03 ± 0.20	42.63 ± 1.13	10.64 ± 0.46	0.35 ± 0.09	8.05 ± 0.03	64.99 ± 1.24	34.14 ± 0.65
3			200	2.76 ± 0.13	42.91 ± 0.64	9.38 ± 0.38	0.46 ± 0.08	8.06 ± 0.04	71.61 ± 1.03	39.90 ± 1.03
4		1:3	120	4.36 ± 0.42	23.57 ± 0.86	8.26 ± 0.11	0.26 ± 0.05	5.91 ± 0.04	68.04 ± 0.50	43.75 ± 0.28
5			160	4.28 ± 0.08	21.74 ± 0.68	8.81 ± 0.22	0.35 ± 0.04	6.59 ± 0.30	69.30 ± 1.04	27.40 ± 0.62
6			200	3.74 ± 1.12	23.59 ± 0.20	8.94 ± 0.16	0.30 ± 0.06	6.70 ± 0.10	73.59 ± 1.62	42.78 ± 0.47
7		3:1	120	5.32 ± 0.48	46.53 ± 1.69	9.60 ± 0.38	0.37 ± 0.03	8.57 ± 0.54	60.89 ± 1.85	27.41 ± 0.22
8			160	4.43 ± 0.86	53.11 ± 0.87	11.36 ± 0.12	0.37 ± 0.05	10.04 ± 0.29	66.29 ± 0.26	35.11 ± 0.63
9			200	2.91 ± 0.56	46.82 ± 1.05	10.61 ± 0.54	0.33 ± 0.05	9.40 ± 0.02	74.51 ± 0.14	57.66 ± 0.68
10	MD:GA	1:1	120	5.48 ± 0.20	55.69 ± 0.04	10.61 ± 0.40	0.39 ± 0.05	16.72 ± 0.53	61.81 ± 0.62	57.67 ± 0.79
11			160	4.04 ± 0.38	53.27 ± 0.44	12.25 ± 0.16	0.42 ± 0.04	10.32 ± 0.03	69.71 ± 0.91	57.68 ± 0.88
12			200	2.70 ± 0.12	58.91 ± 0.28	14.67 ± 0.40	0.33 ± 0.06	12.00 ± 0.04	67.53 ± 0.97	67.29 ± 0.60
13		1:3	120	5.05 ± 0.96	59.75 ± 0.63	15.75 ± 0.55	0.40 ± 0.03	11.94 ± 0.12	60.94 ± 1.01	50.51 ± 0.43
14			160	3.62 ± 0.07	65.16 ± 0.29	16.49 ± 0.30	0.39 ± 0.03	10.15 ± 0.07	66.64 ± 0.57	86.53 ± 0.86
15			200	3.25 ± 0.55	56.10 ± 0.91	16.72 ± 0.14	0.46 ± 0.02	11.31 ± 0.74	72.66 ± 0.95	72.11 ± 0.49
16		3:1	120	4.48 ± 0.77	59.44 ± 0.10	12.08 ± 0.71	0.36 ± 0.04	13.47 ± 0.10	58.43 ± 0.91	52.92 ± 0.56
17			160	3.26 ± 0.87	53.57 ± 0.49	13.85 ± 0.15	0.39 ± 0.04	15.78 ± 0.95	69.08 ± 0.60	67.24 ± 0.01
18			200	1.93 ± 0.73	60.45 ± 0.32	14.73 ± 0.50	0.36 ± 0.01	11.13 ± 0.07	70.15 ± 0.72	67.30 ± 0.43
19	β-CD:GA	1:1	120	5.37 ± 0.34	42.91 ± 0.68	14.70 ± 0.17	0.50 ± 0.03	8.80 ± 0.08	70.16 ± 0.70	34.12 ± 0.91
20			160	4.47 ± 0.19	42.99 ± 0.43	14.76 ± 0.63	0.50 ± 0.01	8.73 ± 0.14	67.98 ± 0.25	42.78 ± 0.51
21			200	2.92 ± 0.58	41.56 ± 0.72	14.28 ± 0.45	0.50 ± 0.02	8.77 ± 0.01	70.58 ± 0.64	52.88 ± 1.20
22		1:3	120	6.91 ± 0.48	60.64 ± 0.53	16.31 ± 0.75	0.59 ± 0.02	10.00 ± 0.11	68.33 ± 0.56	72.14 ± 0.65
23			160	5.22 ± 0.47	56.26 ± 0.93	16.67 ± 0.51	0.50 ± 0.03	10.26 ± 0.01	70.94 ± 0.32	72.11 ± 1.23
24			200	4.10 ± 0.44	55.66 ± 0.93	17.51 ± 0.54	0.50 ± 0.03	10.77 ± 0.17	74.84 ± 0.31	72.16 ± 0.56
25		3:1	120	5.44 ± 0.64	23.33 ± 0.52	9.68 ± 0.36	0.44 ± 0.03	6.49 ± 0.15	70.22 ± 0.67	45.17 ± 0.27
26			160	5.06 ± 0.29	22.97 ± 0.67	10.80 ± 0.45	0.37 ± 0.02	6.19 ± 0.05	73.20 ± 1.01	38.46 ± 0.85
27			200	4.10 ± 0.37	24.24 ± 0.47	11.85 ± 0.29	0.42 ± 0.02	6.17 ± 0.07	73.26 ± 0.56	40.36 ± 1.07
Mean				4.17	45.91	12.55	0.41	9.70	68.49	51.47

MD = maltodextrin, β-CD = β-cyclodextrin, GA = gum arabic. Results are expressed as the mean ± SD.

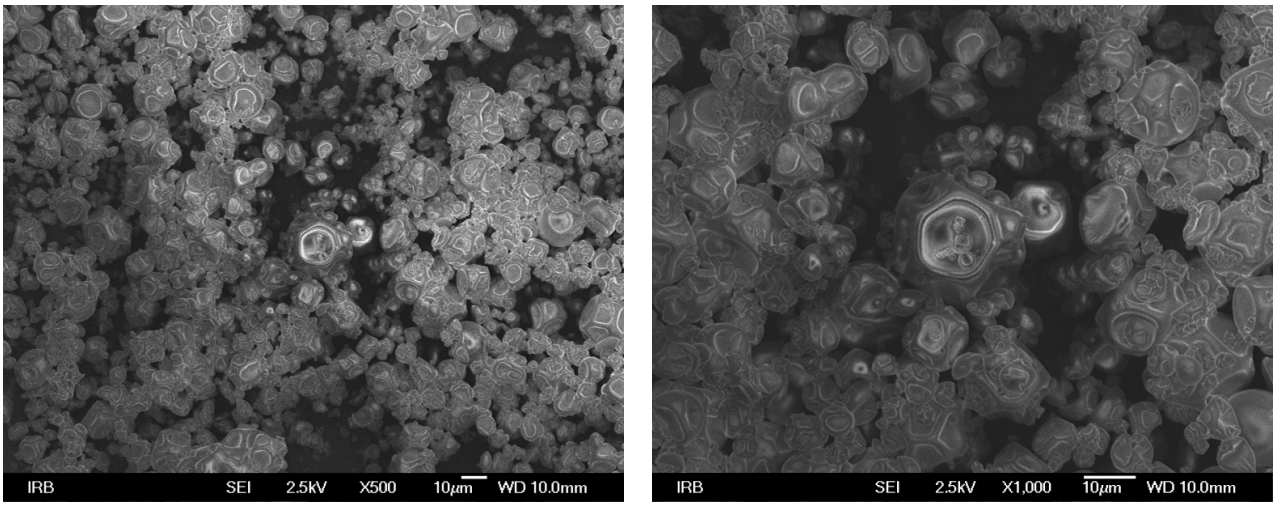
Table 2. Influence of spray-drying conditions on the physical properties of encapsulated fennel essential oil.

Source of Variation	Moisture Content (%)	Solubility (%)	Hygroscopicity (g/100 g)	Bulk Density (g/mL)	Particle Size [d (50), μm]	Powder Recovery (%)	Oil Retention (%)
Wall material type	$p = 0.011$ *	$p < 0.001$ *	$p < 0.001$ *	$p < 0.001$ *	$p < 0.001$ *	$p = 0.005$ *	$p < 0.001$ *
MD:β-CD	3.97 ± 0.23 ^{ab}	38.04 ± 2.71 ^{a}	9.61 ± 0.24 ^{a}	0.35 ± 0.02 ^{a}	7.99 ± 0.31 ^{a}	68.35 ± 1.02 ^{ab}	37.87 ± 2.17 ^{a}
MD:GA	3.76 ± 0.28 ^{a}	58.04 ± 0.87 ^{b}	14.13 ± 0.49 ^{b}	0.39 ± 0.01 ^{a}	12.54 ± 0.54 ^{b}	66.33 ± 1.11 ^{a}	64.36 ± 2.54 ^{b}
β-CD:GA	4.85 ± 0.27 ^{b}	41.17 ± 3.39 ^{a}	14.06 ± 0.63 ^{b}	0.48 ± 0.02 ^{b}	8.46 ± 0.41 ^{a}	71.06 ± 0.54 ^{b}	52.24 ± 3.61 ^{b}
Wall material ratio	$p = 0.363$	$p = 0.402$	$p = 0.107$	$p = 0.094$	$p = 0.976$	$p = 0.321$	$p = 0.024$ *
1:1	3.96 ± 0.26 ^{a}	46.92 ± 1.59 ^{a}	12.24 ± 0.56 ^{a}	0.43 ± 0.02 ^{a}	10.00 ± 0.64 ^{a}	67.81 ± 0.73 ^{a}	46.57 ± 2.89 ^{a}
1:3	4.50 ± 0.27 ^{a}	46.94 ± 4.17 ^{a}	13.94 ± 0.91 ^{a}	0.42 ± 0.03 ^{a}	9.29 ± 0.52 ^{a}	69.48 ± 0.98 ^{a}	59.94 ± 4.43 ^{b}
3:1	4.10 ± 0.29 ^{a}	43.38 ± 3.58 ^{a}	11.62 ± 0.41 ^{a}	0.38 ± 0.01 ^{a}	9.69 ± 0.77 ^{a}	68.45 ± 1.29 ^{a}	47.96 ± 3.24 ^{a}
Drying temperature (°C)	$p < 0.001$ *	$p = 0.867$	$p = 0.293$	$p = 0.940$	$p = 0.962$	$p < 0.001$ *	$p = 0.137$
120	5.25 ± 0.20 ^{c}	45.92 ± 3.36 ^{a}	11.76 ± 0.71 ^{a}	0.41 ± 0.02 ^{a}	10.05 ± 0.79 ^{a}	64.98 ± 1.04 ^{a}	46.26 ± 3.19 ^{a}
160	4.16 ± 0.19 ^{b}	45.74 ± 3.41 ^{a}	12.85 ± 0.63 ^{a}	0.40 ± 0.02 ^{a}	9.57 ± 0.65 ^{a}	68.68 ± 0.59 ^{a}	51.27 ± 4.67 ^{a}
200	3.16 ± 0.19 ^{a}	45.58 ± 3.21 ^{a}	13.19 ± 0.72 ^{a}	0.41 ± 0.02 ^{a}	9.37 ± 0.48 ^{a}	72.08 ± 0.56 ^{b}	56.94 ± 3.09 ^{a}
Wall material type × wall material ratio							
	$p = 0.456$	$p = 0.001$ *	$p = 0.002$ *	$p = 0.040$ *	$p < 0.001$ *	$p = 0.420$	$p = 0.738$
MD:β-CD × 1:1	3.55 ± 0.42 ^{a}	42.33 ± 0.40 ^{ab}	9.62 ± 0.35 ^{ab}	0.40 ± 0.03 ^{b}	8.23 ± 0.11 ^{b}	67.52 ± 1.34 ^{a}	35.57 ± 1.42 ^{a}
MD:β-CD × 1:3	4.13 ± 0.25 ^{a}	22.97 ± 0.44 ^{a}	8.67 ± 0.14 ^{a}	0.30 ± 0.02 ^{a}	6.40 ± 0.17 ^{a}	70.31 ± 1.12 ^{a}	37.97 ± 3.35 ^{a}
MD:β-CD × 3:1	4.22 ± 0.49 ^{a}	48.82 ± 1.41 ^{b}	10.52 ± 0.35 ^{b}	0.36 ± 0.02 ^{ab}	9.34 ± 0.29 ^{c}	67.23 ± 2.53 ^{a}	40.06 ± 5.74 ^{a}
	$p = 0.418$	$p = 0.114$	$p < 0.001$ *	$p = 0.178$	$p = 0.139$	$p = 0.957$	$p = 0.520$
MD:GA × 1:1	4.07 ± 0.51 ^{a}	55.96 ± 1.04 ^{a}	12.51 ± 0.75 ^{a}	0.38 ± 0.02 ^{a}	13.01 ± 1.21 ^{a}	66.35 ± 1.51 ^{a}	60.88 ± 2.04 ^{a}
MD:GA × 1:3	3.97 ± 0.40 ^{a}	60.34 ± 1.68 ^{a}	16.32 ± 0.22 ^{b}	0.41 ± 0.02 ^{a}	11.13 ± 0.36 ^{a}	66.75 ± 2.16 ^{a}	69.72 ± 6.62 ^{a}
MD:GA × 3:1	3.22 ± 0.53 ^{a}	57.82 ± 1.36 ^{a}	13.55 ± 0.52 ^{a}	0.37 ± 0.01 ^{a}	13.46 ± 0.87 ^{a}	65.89 ± 2.38 ^{a}	62.49 ± 3.03 ^{a}
	$p = 0.218$	$p = 0.001$ *	$p < 0.001$ *	$p < 0.001$ *	$p = 0.001$ *	$p = 0.116$	$p = 0.003$ *
β-CD:GA × 1:1	4.26 ± 0.47 ^{a}	42.48 ± 0.35 ^{ab}	14.58 ± 0.17 ^{b}	0.50 ± 0.01 ^{b}	8.77 ± 0.03 ^{ab}	69.57 ± 0.54 ^{a}	43.26 ± 3.44 ^{a}
β-CD:GA × 1:3	5.41 ± 0.54 ^{a}	57.52 ± 1.03 ^{b}	16.83 ± 0.30 ^{c}	0.53 ± 0.02 ^{b}	10.34 ± 0.15 ^{b}	71.37 ± 1.20 ^{a}	72.14 ± 0.27 ^{b}
β-CD:GA × 3:1	4.87 ± 0.29 ^{a}	23.51 ± 0.30 ^{a}	10.78 ± 0.41 ^{a}	0.41 ± 0.01 ^{a}	6.28 ± 0.07 ^{a}	72.23 ± 0.68 ^{a}	41.33 ± 1.29 ^{a}

Table 2. *Cont.*

Source of Variation	Moisture Content (%)	Solubility (%)	Hygroscopicity (g/100 g)	Bulk Density (g/mL)	Particle Size [d (50), μm]	Powder Recovery (%)	Oil Retention (%)
Wall material type × drying temperature (°C)							
	$p = 0.003$ *	$p = 0.960$	$p = 0.062$	$p = 0.929$	$p = 0.795$	$p < 0.001$ *	$p = 0.029$ *
MD:β-CD × 120	4.85 ± 0.21 ^{b}	37.18 ± 4.42 ^{a}	8.90 ± 0.26 ^{a}	0.35 ± 0.03 ^{a}	7.69 ± 0.57 ^{a}	64.96 ± 1.39 ^{a}	34.61 ± 3.05 ^{ab}
MD:β-CD × 160	3.92 ± 0.32 ^{ab}	39.16 ± 5.84 ^{a}	10.27 ± 0.49 ^{a}	0.36 ± 0.02 ^{a}	8.23 ± 0.64 ^{a}	66.86 ± 0.86 ^{a}	32.22 ± 1.55 ^{a}
MD:β-CD × 200	3.14 ± 0.30 ^{a}	37.77 ± 4.55 ^{a}	9.64 ± 0.34 ^{a}	0.36 ± 0.04 ^{a}	8.06 ± 0.49 ^{a}	73.24 ± 0.65 ^{b}	46.78 ± 3.49 ^{b}
	$p < 0.001$ *	$p = 0.532$	$p = 0.095$	$p = 0.826$	$p = 0.069$	$p < 0.001$ *	$p = 0.006$ *
MD:GA × 120	5.00 ± 0.29 ^{c}	58.29 ± 0.83 ^{a}	12.81 ± 0.98 ^{a}	0.38 ± 0.01 ^{a}	14.04 ± 0.90 ^{a}	60.40 ± 0.70 ^{a}	53.70 ± 1.34 ^{a}
MD:GA × 160	3.64 ± 0.22 ^{b}	57.34 ± 2.48 ^{a}	14.20 ± 0.78 ^{a}	0.40 ± 0.01 ^{a}	12.08 ± 1.18 ^{a}	68.48 ± 0.63 ^{b}	70.49 ± 5.37 ^{b}
MD:GA × 200	2.63 ± 0.30 ^{a}	58.49 ± 0.83 ^{a}	15.37 ± 0.44 ^{a}	0.38 ± 0.03 ^{a}	11.48 ± 0.22 ^{a}	70.11 ± 0.98 ^{b}	68.90 ± 1.03 ^{b}
	$p < 0.001$ *	$p = 0.895$	$p = 0.832$	$p = 0.359$	$p = 0.983$	$p = 0.025$ *	$p = 0.755$
β-CD:GA × 120	5.91 ± 0.35 ^{b}	42.29 ± 6.82 ^{a}	13.56 ± 1.27 ^{a}	0.51 ± 0.03 ^{a}	8.43 ± 0.65 ^{a}	69.57 ± 0.44 ^{a}	50.48 ± 7.14 ^{a}
β-CD:GA × 160	4.92 ± 0.18 ^{b}	40.74 ± 6.12 ^{a}	14.08 ± 1.11 ^{a}	0.46 ± 0.03 ^{a}	8.39 ± 0.75 ^{a}	70.71 ± 0.98 ^{ab}	51.12 ± 6.69 ^{a}
β-CD:GA × 200	3.71 ± 0.29 ^{a}	40.49 ± 5.75 ^{a}	14.55 ± 1.05 ^{a}	0.47 ± 0.02 ^{a}	8.57 ± 0.84 ^{a}	72.89 ± 0.80 ^{b}	55.13 ± 5.86 ^{a}

MD = maltodextrin, β-CD = β-cyclodextrin, GA = gum arabic. * $p \leq 0.05$. Results are expressed as the mean ± SE. Values with different letters within each column are statistically different at $p \leq 0.05$.



(**a**) (**b**)

Figure 2. Scanning electron micrographs of the encapsulated fennel essential oil obtained under optimal spray-drying conditions (maltodextrin:gum arabic in a ratio of 1:3 spray-dried at 200 °C) with (**a**) 500× magnification; (**b**) 1000× magnification.

Table 3. Composition (mg/mL) of initial and encapsulated fennel essential oil obtained under optimal spray-drying conditions (maltodextrin:gum arabic in a ratio of 1:3 spray-dried at 200 °C).

No.	Compound	RI	RT	*p*-Value	Initial Fennel EO	Encapsulated Fennel EO
					mg/mL	
	Monoterpene hydrocarbons					
1	α-Pinene	937	5.107	<0.001 *	39.29 ± 0.76 ^{b}	23.17 ± 0.25 ^{a}
2	Camphene	953	5.475	0.001 *	2.08 ± 0.07 ^{b}	1.51 ± 0.09 ^{a}
3	Sabinene	976	6.097	<0.001 *	0.73 ± 0.01 ^{b}	0.52 ± 0.02 ^{a}
4	β-Pinene	980	6.198	<0.001 *	3.32 ± 0.08 ^{b}	2.22 ± 0.02 ^{a}
5	Myrcene	992	6.548	<0.001 *	13.45 ± 0.29 ^{b}	8.44 ± 0.13 ^{a}
6	α-Phellandrene	1006	6.957	<0.001 *	7.73 ± 0.02 ^{b}	5.43 ± 0.05 ^{a}
7	α-Terpinene	1019	7.336	<0.001 *	2.40 ± 0.05 ^{b}	1.40 ± 0.11 ^{a}
8	*p*-Cymene	1027	7.580	<0.001 *	1.66 ± 0.00 ^{b}	1.49 ± 0.03 ^{a}
9	D-Limonene	1031	7.710	<0.001 *	25.46 ± 0.18 ^{b}	17.46 ± 0.16 ^{a}
11	γ-Terpinene	1062	8.712	<0.001 *	2.29 ± 0.02 ^{b}	1.94 ± 0.03 ^{a}
12	*cis*-Sabinene hydrate	1069	8.997	0.006 *	0.44 ± 0.02 ^{b}	0.37 ± 0.00 ^{a}
	Oxygenated monoterpenes					
10	Eucalyptol	1035	7.817	0.002 *	0.92 ± 0.06 ^{b}	0.63 ± 0.05 ^{a}
13	L-Fenchone	1090	9.767	<0.001 *	170.82 ± 0.45 ^{b}	134.69 ± 0.33 ^{a}
14	Linalool	1100	10.176	0.001 *	2.05 ± 0.05 ^{b}	1.65 ± 0.04 ^{a}
15	Camphor	1147	11.884	<0.001 *	2.44 ± 0.02 ^{b}	1.94 ± 0.04 ^{a}
16	α-Terpineol	1179	13.194	<0.001 *	2.27 ± 0.01 ^{b}	1.95 ± 0.02 ^{a}
18	Carvone	1244	15.827	0.236	1.15 ± 0.08 ^{a}	1.08 ± 0.01 ^{a}
	Phenylpropanoids					
17	Estragole	1198	14.066	<0.001 *	36.42 ± 0.25 ^{b}	29.57 ± 0.09 ^{a}
20	*trans*-Anethole	1288	17.771	<0.001 *	659.99 ± 2.87 ^{b}	564.92 ± 3.64 ^{a}
	Others					
19	*p*-Anisaldehyde	1256	16.325	<0.001 *	15.58 ± 0.12 ^{b}	11.46 ± 0.16 ^{a}
	Total (%)	*Monoterpene hydrocarbons*		<0.001 *	9.98 ± 0.14 ^{b}	7.88 ± 0.10 ^{a}
		Oxygenated monoterpenes		<0.001 *	18.14 ± 0.05 ^{b}	17.48 ± 0.09 ^{a}
		Phenylpropanoids		<0.001 *	70.31 ± 0.18 ^{a}	73.23 ± 0.21 ^{b}
		Others		<0.001 *	1.57 ± 0.01 ^{b}	1.41 ± 0.02 ^{a}

EO = essential oil. Results are expressed as the mean ± SD. * $p \leq 0.05$. Values with different letters within each row are statistically different at $p \leq 0.05$.

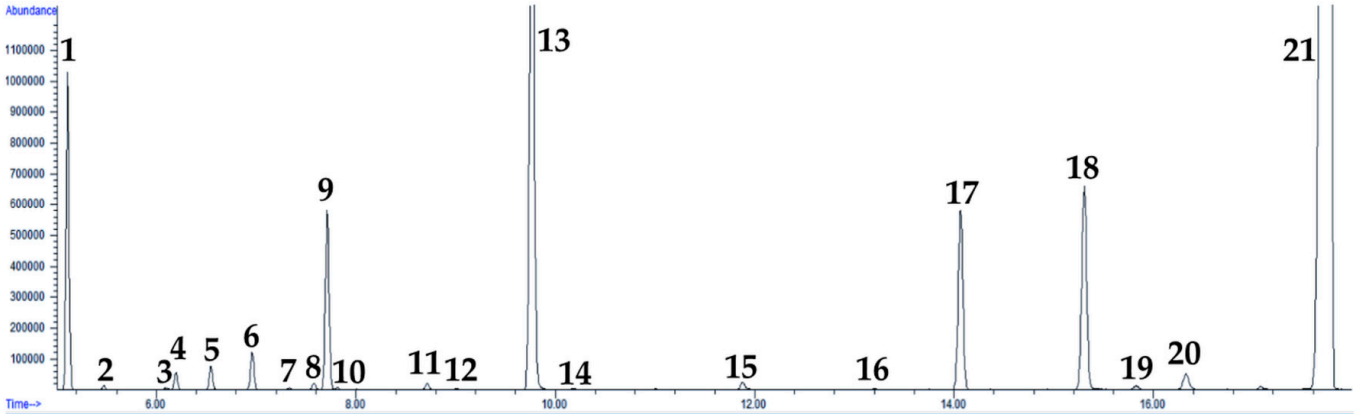


Figure 3. GC-MS chromatogram of the initial fennel essential oil (1 = α-Pinene, 2 = Camphene, 3 = Sabinene, 4 = β-Pinene, 5 = Myrcene, 6 = α-Phellandrene, 7 = α-Terpinene, 8 = *p*-Cymene, 9 = D-Limonene, 10 = Eucalyptol, 11 = γ-Terpinene, 12 = *cis*-Sabinene hydrate, 13 = L-Fenchone, 14 = Linalool, 15 = Camphor, 16 = α-terpineol, 17 = Estragole, 18 = Nerol (IS), 19 = Carvone, 20 = *p*-Anisaldehyde, 21 = *trans*-Anethole).

3.1. Physical Properties of Encapsulated Fennel Essential Oil

3.1.1. Moisture Content

The moisture content is one of the most important parameters for the quality and stability of the produced powder. It is desirable that it does not exceed 5% in order to create unfavorable conditions for microbial growth and better overall stability, which allows a longer shelf life and possible use for further purposes [40]. The moisture content of the fennel EO powders was determined in the range of 1.93–6.91% with a mean value of 4.17% (Table 1), with most powders meeting the moisture content criteria. The obtained values are in accordance with the findings of Felix et al. [15] and Alvarenga Botrel et al. [14] who spray-dried cinnamon and oregano EO, respectively, while Fernandes et al. [12] and Marques et al. [13] reported a slightly lower span of moisture content in rosemary (0.26–3.16%) and thyme (1.32–2.06%) EO powders.

The statistical results showed that the moisture content of the powders was significantly influenced by the type of wall material and the drying temperature, while the wall material ratio had no significant influence (Table 2). Regarding the wall material type, powders prepared with MD:GA had the lowest moisture content (mean value 3.76%), while the highest moisture content was present in β-CD:GA powders (mean value 4.85%). Powders containing MD:β-CD had a similar moisture content (mean value 3.97%) to MD:GA powders. This clearly shows that the presence of MD contributes to the most effective production of spray-dried fennel EO powders when it comes to the wall material choice. MD is soluble in water and has a good membrane-forming capability due to its medium molecular weight, which suggests its application in encapsulation to extend the storage time of the encapsulated product [17], while the molecular structure of GA and β-CD could hinder molecular diffusion during the spray-drying process [12]. A similar effect in increasing the moisture content in spray-dried powders when GA and β-CD were incorporated as wall materials has also been previously documented [41–43]. Although it was not statistically significant parameter, the obtained results for the wall material ratio showed that a ratio 1:1 contributed to the lowest moisture content. Furthermore, increasing the drying temperature led to a significant decrease in moisture content in a linear trend, with the lowest moisture content being reached at the highest temperature (200 °C). This was to be expected, as the application of a higher temperature increases the mass and heat transfer, leading to rapid evaporation and thus to a rapid removal of moisture. Alvarenga Botrel et al. [14] also reported the lowest moisture content in oregano EO powders produced around the maximum of the tested temperature range (132–188 °C), as did Fernandes et al. [12] in

rosemary EO powders obtained at the highest applied temperature (135–195 °C). The same results were also documented by authors who spray-dried plant extracts [6,41].

3.1.2. Solubility

In addition to the moisture content, the solubility of the powder is also an essential parameter which describes the ability of the powder to redissolve and transform into a liquid form for further processing and use, whereby a lack of solubility can lead to complications [44]. The solubility of powders is influenced by the moisture content and particle size, with a low moisture content increasing solubility and larger particles sinking and dissolving more quickly [6]. The solubility of the fennel EO powders ranged from 21.74 to 65.16% with a mean value of 45.91% (Table 1), with most of the fennel EO powders exhibiting good solubility despite the hydrophobic character of the core material. The results obtained in this study are in line with the results obtained by Felix et al. [15] and Fernandes et al. [11] for the solubility of cinnamon and rosemary EO powders (33.04–49.57% and 55.75–67.75%, respectively), but slightly lower than the values obtained by Marques et al. [13] for thyme EO powders (64.09–70.30 g/100 g). However, the latter authors used different wall materials, namely whey protein isolate and MD, both alone and in combination, as well as with the addition of chitosan.

The solubility of fennel EO powders was only significantly influenced by the choice of wall material, while the wall material ratio and drying temperature did not cause any significant differences in the solubility of the powders. When considering the influence of the type of wall material, the highest solubility value was found for MD:GA powders (mean value 58.04%), while wall material combinations containing β-CD had a lower solubility (β-CD:GA mean value 41.17%, MD:β-CD mean value 38.04%). In comparison with other studies, the findings in this study are generally consistent with reported data, where Fernandes et al. [11] and Felix et al. [15] reported a solubility of 45.82% for rosemary EO powder and 37.38% for cinnamon EO powder, both containing MD:GA in a 1:1 ratio. Moreover, other authors [6,7,41,42,45] also confirmed decreased values for this reconstitution property when β-CD was included as the wall material. A mixture of MD and GA has already shown good solubility properties [46], for which the chemical constitution of the two wall materials is responsible. As already mentioned, MD contains numerous hydroxyl groups, which facilitate the dissolution process, while GA has a highly branched structure and good emulsifying properties, which favors good solubility. On the other hand, β-CD is the least water-soluble of the wall materials used due to the intramolecular hydrogen bonds between the hydroxyl groups of neighboring glucose units [6]. The solubility of the fennel EO powders was quite similar regardless of the ratio of the wall materials (approximately 46%) and the drying temperature (approximately 45%). Fernandes et al. [12] also found no significant effect of drying temperature on the solubility of rosemary EO powders, but they only investigated GA as a wall material.

3.1.3. Hygroscopicity

The hygroscopicity of powders describes their behavior in a high-humidity environment and their ability to absorb moisture, respectively. It is very important to determine this parameter in terms of powders' stability. In addition, hygroscopicity, together with solubility, has a major influence on the reconstitution of powders, which is very important in the food industry, especially when encapsulated flavors and oils are to be applied to aqueous products [14]. Therefore, reduced hygroscopicity is desirable, as moisture can lead to agglomeration. The results for the hygroscopicity of the fennel EO powders after 7 days showed that it ranged from 8.26 to 17.51 g/100 g, with a mean value of 12.55 g/100 g (Table 1). According to Pisecky [47], powders can be categorized by their hygroscopicity as hygroscopic (15–20%), slightly hygroscopic (10–15%) and non-hygroscopic powders (<10%). Hence, the majority of the fennel EO powders prepared in this study can be classified as slightly hygroscopic to non-hygroscopic. The results obtained are in relatively good agreement with some literature data on the hygroscopicity of EO powders [12,13],

but some authors reported higher hygroscopicity values, e.g., 22.30–26.27% by Alvarenga Botrel et al. [14] for oregano EO powders and even 22.9–42.21% by Felix et al. [15] for cinnamon EO powders.

As for the influence of the individual factors tested, the results showed that the type of wall material was the only factor with a significant influence, while neither the wall material ratio nor the drying temperature had a significant effect on the hygroscopicity of the fennel EO powders (Table 2). The lowest hygroscopicity was found for MD:β-CD powders (mean value 9.61 g/100 g), while the powders produced from the other two wall materials showed significantly higher values for hygroscopicity (mean value 14.13 and 14.06 g/100 g, respectively). A similar observation about the low hygroscopicity of powders containing MD:β-CD was also made by Dobroslavić et al. [7] who spray-dried laurel extract. Furthermore, in their study, powders consisting of β-CD:GA were also more hygroscopic, as in this study. This is related to the specific structure of the wall materials used, as β-CD consists of an inner hydrophobic cavity and a hydrophilic outer part and is therefore less inclined to bind water molecules, while MD itself is less hygroscopic and its lower polymerization degree leads to a lower degree of water adsorption. Conversely, a highly branched form of GA causes water molecules to bind to the hydroxyl groups in the chains.

3.1.4. Bulk Density

From an economic and functional point of view, the bulk density of powders is a decisive characteristic for processing, packaging, storage and distribution. A low bulk density is unfavorable, as it not only leads to higher packaging costs, but also accumulates a greater amount of air between the particles, which increases the possibility of oxidation of the product and thus reduces storage stability [11]. The values obtained for the bulk density of fennel EO powders ranged from 0.26 to 0.59 g/mL, with 0.41 g/mL being the mean value (Table 1). These results are consistent with previous studies on spray-dried EO [11–15,46].

The bulk density of the EO powders was significantly affected by the type of wall material, while the ratio of wall materials and the drying temperature had no significant effect (Table 2). The highest value of bulk density was determined for powders prepared with β-CD:GA (mean value 0.48 g/mL), while powders containing combinations with MD had a lower and quite similar bulk density (MD:β-CD mean value 0.35 g/mL, MD:GA mean value 0.39 g/mL). Marques at al. [13] also confirmed that the choice of wall material has a direct influence on the distribution and size of the powder particles. They also reported the decrease in bulk density due to the presence of MD in a mixture of MD and whey protein isolate compared to whey protein isolate alone. In another study, bulk density was found to decrease with increasing MD concentration [46]. The results of that study showed that powders prepared with MD DE 6, which is similar to the MD used in this study, had the lowest bulk density, in contrast to powders containing MD with a higher DE. The authors explained this behavior with the lower moisture content or higher air entrapment in the particles, as MD is a skin-forming material. Furthermore, the higher DE of MD reflects its lower glass transition temperature, making the mixture stickier. In addition, Cai and Corke [48] also stated that air entrapment in the particles can cause low bulk density.

3.1.5. Particle Size

Particle size is of great importance in spray-drying, as it can influence not only the appearance and density of the powder, but also the dispersion of powders in liquids [49]. Although powders with larger particles are difficult to disperse in final products, larger particles generally protect the core material better than smaller particles. The particle size of the produced fennel EO powders, expressed as the d (50) value, was determined in the range of 5.91–16.72 μm, where 9.70 μm was the mean value (Table 1). The determined particle size range of fennel EO powders is in agreement with the results of other studies

on EO powders [11,15,18]. According to Ré [50], encapsulated particles can be categorized into three groups based on their size: macro- (>5000 μm), micro- (0.2–5000 μm) and nanoparticles (<0.2 μm). Based on this classification, the fennel EO produced in this study can be considered as microparticles. The results obtained show a broad particle size distribution, which according to Costa et al. [25] is a regular characteristic of spray-dried particles. Moreover, Goula and Adamopoulos [51] have stated that larger particles generally have a lower density, so that the bulk density of a powder with a large particle size is lower. This relationship between particle size and bulk density is generally visible in the behavior of the fennel EO powders, as the MD:GA powders, for example, were characterized by the largest particle size and a lower bulk density (Table 2).

Again, the wall material type was the only significant factor influencing particle size (Table 2). MD:β-CD and β-CD:GA powders had a smaller particle size (mean values 7.99 and 8.46 μm, respectively), in contrast to MD:GA powders, which were characterized by significantly larger particles (mean value 12.54 μm). This can be attributed to the presence of GA and its higher viscosity, since a higher viscosity of the wall material results in a higher viscosity of the feed solution and correspondingly larger droplets are formed during atomization [52]. The low viscosity of MD at high concentrations contributes to the formation of smaller particles. Fernandes et al. [11] noticed a similar behavior of GA in their study. They used different wall materials in their study, including GA alone and in combination with MA. They found that the use of GA alone resulted in the largest particles of rosemary EO powder, while the addition of MD to GA decreased the particle size of the EO powder. A study by Felix et al. [15] also showed that the particle size of cinnamon EO powders decreased when MD was combined with GA, as opposed to using GA alone.

3.1.6. Powder Recovery

Along with the physical properties of the powders produced, the powder recovery, i.e., the process yield, is of crucial importance from an economic point of view as it gives an insight about the efficiency of the spray-drying process itself. The unwanted loss of particles leading to reduced powder recovery, can be caused by improper spray-drying conditions, e.g., sticking to the wall of the drying chamber or loss through an exhaust air filter or during manual removal of the powder from the collection container. The results obtained for the powder recovery for the spray-drying of fennel EO are shown in Table 1. The powder recovery ranged from 58.43 to 74.84%, with a mean value of 68.49%, which can be considered as a satisfactory process yield [53]. In comparison, other authors [14,18,42] reported a slightly lower range for the powder recovery of different EO powders, which could be due to different experimental designs.

The powder recovery was significantly influenced by the type of wall material and the drying temperature, while the wall material ratio had no significant effect on this property (Table 2). The highest powder recovery was obtained in the presence of β-CD:GA (mean value 71.06%), followed by MD:β-CD (mean value 68.35%), while the lowest yield was present when MD:GA was used as the wall material (mean value 66.33%). Dobroslavić et al. [7] also documented a high powder recovery (> 70%) for powders of laurel extract prepared by combining β-CD with MD or GA. Čulina et al. [45] who spray-dried sea buckthorn berry oil also confirmed the highest values of powder recovery obtained when using β-CD:GA (50.71–65.61%).

The effect of temperature showed a linear trend where the powder recovery increased with increasing drying temperature, i.e., at the highest applied temperature (200 °C) the highest amount of powder (mean value 72.08%) was obtained. The temperatures of 120 and 160 °C yielded 64.98 and 68.68% (mean values) of the powder, and these amounts did not differ significantly (Table 2). According to Da Veiga et al. [20], increasing the temperature facilitates the drying process and reduces the surface tension and viscosity, which contributes to the formation of droplets leading to the greater efficacy of the spray-drying process. The same effect of drying temperature increase on the powder recovery was noticed by Alvarenga Botrel et al. [14] who varied the drying temperature in the

spray-drying of oregano EO in the range of 132–188 °C, and Čulina et al. [45] who used 120, 150 and 180 °C as the drying temperature in the production of sea buckthorn berry oil powders.

3.1.7. Oil Retention

Another quality parameter for estimating the spray-drying efficiency is certainly the oil retention, i.e., the percentage of the initial amount of EO that is encapsulated [11]. A higher value of oil retention indicates a successful EO encapsulation. The oil retention in the fennel EO powders ranged from 27.40 to 86.53% with a mean value of 51.47% (Table 1), which can be considered sufficient. Among other requirements, the use of the emulsifier Tween 20 provided this beneficial outcome, as it enables a lower interfacial tension, which leads to a better interaction between the wall material and the EO and thus allows a higher oil retention [20]. The determined retention of fennel EO is consistent with literature data [11,16,25], although the upper range of the values determined in this study is slightly higher than the values reported in the listed studies (60.22–77.39%). On the other hand, Alvarenga Botrel et al. [14] reported notably lower values for the oil retention of oregano EO (5.1–33.9%), but they used a lower concentration of the wall material.

Statistical analysis showed a significant influence of the type of wall material and wall material ratio on oil retention in fennel EO powders, while the powders did not differ significantly in the oil retention values in relation to the drying temperature (Table 2). Regarding the wall material type, the highest oil retention was achieved when using MD:GA (mean value 64.36%), followed by β-CD:GA (mean value 52.24%), while the presence of MD:β-CD contributed the least to the oil retention (mean value 37.87%). The results show that volatiles were more strongly retained in the mixtures containing GA, which is to be expected due to the excellent emulsifying properties of GA, which consequently favor the retention of volatiles during spray-drying, as the better stability of the feed solution results in a higher encapsulation efficiency and reduced loss of volatiles, respectively. Fernandes et al. [11] and Thuong Nhan et al. [17] reported a slightly lower value for the retention of rosemary (45.45%) and lemongrass EO (54.88%) in powders containing the same amount of MD:GA as in this study (20%).

With respect to the wall material ratio, the highest oil retention was obtained when a wall material ratio of 1:3 was used (mean value 59.94%), while ratios of 1:1 and 3:1 resulted in significantly lower oil retention (mean values 46.57 and 47.96%, respectively).

3.1.8. PCA Analysis

PCA analysis was performed on the physical properties of the fennel EO powders to examine the relationship between the powders and to test their possible grouping according to the factors tested (type of wall material, ratio of wall materials and drying temperature). The PCA showed the grouping of the powder samples only in relation to the type of wall material (Figure 1), while neither the wall material ratio nor the drying temperature allowed separation of the powder samples, which is why these results are not presented. This confirms that the type of wall material was the most important influencing factor, as discussed earlier.

Based on the preliminary communality test, only solubility, hygroscopicity, particle size, powder recovery and oil retention were included in the analysis as their communality value was >0.5. PC1 and PC2 accounted for 86.74% of the total data variance. PC1 correlated very strongly ($r > 0.83$) with all parameters included in the test, with the exception of powder recovery ($r = 0.24$). As for PC2, there was a strong correlation ($r = -0.93$) only between PC2 and powder recovery, while hygroscopicity and oil retention correlated moderately with this PC ($r > 0.40$). The correlation between particle size and PC2 was moderate ($r = 0.35$), and solubility correlated very weakly ($r = 0.17$). As it can be seen in Figure 1, PCA showed a clear separation between the MD:β-CD and MD:GA powders. All MD:β-CD powders were situated at the positive values of PC1, as they were characterized by relatively high values for powder recovery, contrarily to the MD:GA powders, which took place at negative

values of PC1, completely separating from the MD:β-CD powders by higher values for particle size, solubility, oil retention and hygroscopicity. The β-CD:GA powders clustered at the negative values of PC2 and exhibited higher powder recovery, oil retention and hygroscopicity.

Finally, oil retention was selected as the main criterion for determining the optimal spray-drying conditions for fennel EO, as this parameter determines the quality of the final product. Therefore, a combination of MD:GA at a ratio of 1:3 and spray-dried at 200 °C can be defined as the optimal conditions for spray-drying fennel EO (Table 2). The fennel EO powder obtained under these conditions can be considered as the one having the best prospective process outcomes in terms of high powder recovery (72.66%) and oil retention (72.11%), together with the desirable characteristics of low moisture content (3.25%) and high solubility (56.10%) (Table 1).

3.1.9. SEM Analysis

The morphology of the particles of fennel EO powder obtained under the optimal spray-drying conditions was analyzed by SEM at 500× and 1000× magnification (Figure 2). The particles were of relatively uniform morphology and did not show cracks, indicating successful encapsulation. Most of the particles had a semi-spherical shape with a slightly shriveled and concave outer surface. Similar structures were observed by Felix et al. [15] for the MD:GA powder of cinnamon EO. According to Ré [50], spray-dried particles are usually hollow spheres that are formed by the shrinkage after the outer surface hardens and the subsequent expansion of the air bubbles captured inside the droplet.

3.2. Chemical Composition of Initial and Encapsulated Fennel Essential Oil

To assess the quality of the encapsulated fennel EO in terms of its chemical composition, the initial fennel EO and the one hydrodistilled from the EO powder prepared under optimal conditions (MD:GA/1:3/200 °C) were analyzed by GC-MS. The comparison of their chemical profiles is shown in Table 3.

GC-MS analysis detected a total of 20 compounds (α-pinene, camphene, sabinene, β-pinene, myrcene, α-phellandrene, α-terpinene, *p*-cymene, D-limonene, eucalyptol, γ-terpinene, *cis*-sabinene hydrate, L-fenchone, linalool, camphor, α-terpineol, estragole, carvone, *p*-anisaldehyde and *trans*-anethole) (Figure 3) belonging to monoterpene hydrocarbons (11), oxygenated monoterpenes (6), phenylpropanoids (2) and aromatic aldehydes (others) (1) in both oils tested, so that spray-drying showed no negative influence on the qualitative nature of the fennel EO. Both oils were characterized by the same distribution of chemical groups: phenylpropanoids were the most abundant (70.31 and 73.23%), followed by oxygenated monoterpenes (18.14 and 17.48%) and monoterpene hydrocarbons (9.98 and 7.88%), while aromatic aldehydes were the least represented (1.57 and 1.41%). A similar composition of fennel EO was also reported by Marčac et al. [1] who analyzed fennel EO obtained by hydrodistillation and steam distillation with or without pretreatment by cryogrinding. Of the compounds identified, α-pinene, myrcene, D-limonene, L-fenchone, estragole, *trans*-anethole and *p*-anisaldehyde were the most abundant in both oils with a concentration > 8 mg/mL. *Trans*-anethole was found as the main constituent with a concentration of 659.99 and 564.92 mg/mL in initial and encapsulated fennel EO. This was also confirmed in other studies [1,54–56]. *Trans*-anethole is known as one of the aroma carriers of fennel EO responsible for its characteristic flavor [57]. L-fenchone was the second most abundant, present at concentrations of 170.82 and 134.69 mg/mL, while α-pinene and estragole were present in similar concentrations (39.29 and 36.42 mg/mL in the initial EO and 23.17 and 29.57 mg/mL in the encapsulated EO, respectively), as were myrcene and *p*-anisaldehyde (13.45 and 15.58 mg/mL in the initial EO and 8.44 and 11.46 mg/mL in the encapsulated EO, respectively).

Regarding the quantitative aspect, the results revealed significant differences in the amounts of the components of the initial and encapsulated EO. It was found that the concentrations of all compounds were significantly lower in the encapsulated EO compared

to the initial EO, with the exception of carvone, the amounts of which were not significantly different in both oils. This was to be expected due to the volatile nature of the EO components and their easy possible degradation by the heat applied or by oxidation in the presence of air during atomization [58]. According to King [59], the loss of volatiles during spray-drying can generally be caused by the following factors: (i) facilitated volatilization due to the large surface area during atomization; (ii) facilitated diffusion of volatiles when the membrane of the particles does not form quickly; (iii) leakage of volatiles due to vapor bubbles inside the particles. However, it should be noted that the average loss of volatiles was 24.2%, which cannot be considered as inadequate. The greatest loss was observed for α-pinene and α-terpinene (approximately 40%) and for most monoterpene hydrocarbons (approximately 30%), which are highly volatile. Due to their low molecular weight, the monoterpenes tend to escape from the droplets at higher temperatures [17]. Moreover, Marques et al. [13] explained that the loss of monoterpene hydrocarbons is related to their hydrophobicity, due to which they are more concentrated on the surface of the particles and therefore less protected and more prone to volatilization. The concentrations of other compounds decreased by about 15–20%, and carvone showed its good stability (decrease of about 6%). These changes in the quantitative composition of the encapsulated EO are reflected in the relative amount of chemical groups, where on account of the significant decrease in monoterpene hydrocarbons, oxygenated monoterpenes and aromatic aldehydes, the level of phenylpropanoids rose markedly. The changes in the chemical composition of the EO prior to and after the spray-drying were also reported by other authors [11,13,18]. They also confirmed a loss of volatiles due to spray-drying, but with no or only minimal changes in the qualitative structure and the altered relative amount of compounds.

However, despite the observed loss of volatiles, it can be concluded that the encapsulated EO retained its quality, proving that the encapsulation of fennel EO by spray-drying did not compromise it and was quite successful under the given conditions. In addition, the authors themselves testified to the intense and pleasant characteristic fennel aroma of the fennel EO powder produced, which also indicates effective encapsulation of the fennel EO.

4. Conclusions

In this study, the encapsulation of fennel essential oil was successfully performed using the spray-drying method. All obtained powders generally exhibited good characteristics, with an emphasis on the low moisture content, high powder recovery and high oil retention. Of all the effects studied, the type of wall material was the factor that most strongly influenced all properties of the powders, while the drying temperature showed a moderate effect. In contrast, the ratio of wall materials in the examined mixtures only had an influence on the retention of the encapsulated essential oil. The use of all wall material mixtures showed relatively good results, but the MD:GA blend proved to be the most prosperous in achieving favorable physical characteristics in spray-dried powders of fennel essential oil. Together with this choice of wall material, a wall material ratio of 1:3 and a drying temperature of 200 °C were found to be optimal for the spray-drying of fennel essential oil. The powder produced under these conditions was characterized by a low moisture content (3.25%) and high solubility (56.10%), while achieving satisfactory process features such as high powder recovery (72.66%) and oil retention (72.11%). The comparison of the initial and the encapsulated fennel essential oil showed quantitative differences in the chemical composition, but without qualitative changes. The concentrations of the individual volatiles, especially the monoterpene hydrocarbons, decreased in the encapsulated essential oil; however it was not inadequate (average decrease was 24.2%). Finally, the encapsulation of fennel essential oil by spray-drying proved to be a perspective mechanism for its stabilization, expanding the possibilities for its further use in the food, cosmetic and pharmaceutical sectors to improve the original food flavor, as a food preservative or for medicinal and therapeutic purposes. However, future research should target the practical

application of encapsulated fennel essential oil to evaluate its effect, bioavailability and controlled release in different matrices.

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