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Optimization of drying parameters in the microencapsulation of volatile oil from *Spiranthera odoratissima* leaves

Otimização dos parâmetros de secagem na microencapsulação do óleo volátil de folhas de *Spiranthera odoratissima*

Optimización de los parámetros de secado en la microencapsulación de aceite volátil de hojas de *Spiranthera odoratissima*

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Abstract
Spiranthera odoratissima A. St.-Hil. (Rutaceae), *"manacá"*, is popularly used for head, muscle aches, rheumatism and, stomach, uterine, renal and liver disorders. The aims of this study were to investigate the physico-chemical and morphological properties of microencapsulated powder of volatile oil from *S. odoratissima* leaves, optimize the drying process and verify the influence of drying parameters on microencapsulation by spray-drying. The volatile oils from leaves were extracting by hydrodistillation in a Clevenger type apparatus and analyzed by GC/MS. The emulsions were prepared and spray-dried. Box-Behnken experimental model was used for optimize the effects of drying parameters on the encapsulation responses. The β-caryophyllene content in the microcapsules was determined by HPLC. The results suggest that the best operational conditions for the atomization drying of *S. odoratissima* volatile oil were inlet temperature of 158°C, feed flow of 0.25L/h and drying nozzle diameter of 0.7mm. These results reveal the technological potential of the microcapsules obtained from *S. odoratissima* volatile oils.
Keywords: Wall materials; Box-behnken design; β-caryophyllene; GC-MS; “Manacá”; Rutaceae.

Resumo

Spiranthera odoratissima A. St.-Hil. (Rutaceae), "manacá", é utilizada popularmente para tratar dores de cabeça, de estômago, doenças musculares, reumatismo, disordens do útero, rins e fígado. Os objetivos deste estudo foram investigar as propriedades físico-químicas e morfológicas do pó microencapsulado do óleo volátil das folhas de *S. odoratissima*, otimizar o processo de secagem e verificar a influência dos parâmetros de secagem na microencapsulação por spray-dry. Os óleos voláteis das folhas foram extraídos por hidrodestilação em aparelho tipo Clevenger e analisados por CG / EM. As emulsões foram preparadas e secas por pulverização. O modelo experimental Box-Behnken foi utilizado para otimizar os efeitos dos parâmetros de secagem nas respostas de encapsulamento. O conteúdo de β-cariofileno nas microcápsulas foi determinado por HPLC. Os resultados sugerem que as melhores condições operacionais para a secagem por atomização do óleo volátil de *S. odoratissima* foram temperatura de entrada de 158 °C, vazão de alimentação de 0,25L/ h, diâmetro do bico de secagem de 0,7mm. Esses resultados revelam o potencial tecnológico das microcápsulas obtidas a partir de óleos voláteis de *S. odoratissima*.

Palavras-chave: Materiais de parede; Desenho de Box-behnken; β-cariofileno; GC-MS; "Manacá"; Rutaceae.

Resumen

Spiranthera odoratissima A. St.-Hil. (Rutaceae), "manacá", se usa popularmente para dolores de cabeza, musculares, reumatismo, estomago, útero, renales y trastornos hepáticos. Los objetivos de este estudio fueron investigar las propiedades físico-químicas y morfológicas del polvo microencapsulado de aceite volátil de hojas de S. odoratissima, optimizar el proceso de secado y verificar la influencia de los parámetros de secado en la microencapsulación por atomización. Los aceites volátiles de las hojas se extrajeron por hidrodestilación en un aparato tipo Clevenger y se analizaron por GC / MS. Las emulsiones se prepararon y se secaron por pulverización. Se utilizó el modelo experimental Box-Behnken para optimizar los efectos de los parámetros de secado en las respuestas de encapsulación. El contenido de β-cariofileno en las microcápsulas se determinó mediante HPLC. Los resultados sugieren que las mejores condiciones operativas para el secado por atomización del aceite volátil de *S. odoratissima* fueron una temperatura de entrada de 158 °C, un flujo de alimentación de 0,25 L / h, un diámetro de boquilla de secado de 0,7 mm. Estos resultados revelan el potencial tecnológico de las microcápsulas obtenidas a partir de aceites volátiles de *S. odoratissima*.

Palabras clave: Materiales de pared; Diseño Box-behnken; β-cariofileno; GC-MS; "Manacá'; Rutaceae.

1. Introduction

Spiranthera odoratissima A. St. –Hil. (Rutaceae), popularly known as "manacá", is a shrub found in the Brazilian Cerrado (Barbosa, et al., 2012; Chaibub, et al., 2013; Cornelio, et al., 2017). The *S. odoratissima* roots is popularly used in the form of tea for stomach pain, headaches, muscle aches, liver disorders and rheumatism, while the tea from the leaves is used for stomach, uterine, kidney and liver disorders (Silva & Felfili, 2012; Trezenzol, et al., 2006; Verde, et al., 2003). Its leaves have secretory cavities containing volatile oil (Jarald, et al., 2008; Matos, et al., 2014).

Scientific studies verified analgesic and anti-inflammatory activities of ethanol extract (Matos, et al., 2003) and anxiolytic activity of volatile oil (VO) from *S. odoratissima* leaves in rats (Galdino, et al., 2012, Oliveira, et al., 2020). The antimicrobial activity of leaf volatile oil was observed against *Micrococcus roseus* 1740 (MIC=0.195mg/ml) and *Candida albicans* NTC 2010 (MIC=1.562 mg/ml) (Chaibub, et al., 2013). Souza et al., (2020) verified antibacterial activity of the most polar extracts from *S. odoratissima* leaves against *L. monocytogenes* (MIC values ranged between 12.5 and 62.5 μg mL).

Chaibub, et al. (2013) identified as major compounds in the VO of *S. odoratissima* leaves collected in Senador Canedo, Goiás, β-caryophyllene (20.64%), γ-muurolene (17.7%), bicyclogermacrene (14.73%) and δ-cadinene (13.40%). Souza, et al. (2018) identified as major compounds in the VO of *S. odoratissima* leaves, collected in Aparecida de Goiânia, Goiás, bicyclogermacrene (17.61%-23.08%), δ-cadinene (12.31%-16.55%), amorphous-4,7(11)-diene (10.71%-19.87%) and β-caryophyllene (6.78%-12.15%) and verified that the β-caryophyllene content did not undergo significant changes in a seasonality study.

Volatile oil may undergo degradation and/or volatilize when exposed to environmental agents such as heat, light, moisture and oxygen (Bakry, et al., 2016; Turasan, et al., 2015). Due to the increasing demand for volatile oils in the pharmaceutical, food (Subtil, et al., 2014) and cosmetics industries, it is interesting to develop technological strategies such as

microencapsulation (ME), which are able to maintain their chemical properties in face of the mentioned factors previously (Bakry, et al., 2016; Rosas, et al., 2017).

The microencapsulation by spray-drying has the advantages of low cost, good volatiles retention and stability of the finished product, besides the flexibility and fastness of the process (Reineccius, 2004). This technique guarantees a satisfactory yield in the drying process, high efficiency encapsulation (EE), high content of microencapsulated material, regardless of the capacity of the equipment and heat sensitivity (Marques, et al., 2016) and biological and pharmacological potential of volatile oils (Asbahani, et al., 2015; Asensio, et al., 2017).

The properties of wall materials, formulation parameters and drying conditions involving the ME process can affect the final characteristics of microencapsulated products (Ribeiro, et al., 2015). These are important conditions in the spray-drying drying process: the air inlet temperature (Frascareli, et al., 2012), the feed flow (Pires & Pena, 2017) and the diameter of the spray nozzle (Alves, et al., 2014). The use of response surface methodology (RSM) allows to verify the effect of the drying parameters on the properties of the obtained material (Baş & Boyaci, 2007).

The aims of this study were to investigate the physico-chemical and morphological properties of microencapsulated powder of volatile oil from *S. odoratissima* leaves, optimize the drying process and verify the influence of drying parameters on microencapsulation by spray-drying.

2. Methodology

2.1 Pant material

The *Spiranthera odoratissima* A. St.-Hil. (Rutaceae) leaves were collected in 2016 (November and December) from approximately 50 individuals, in Aparecida de Goiânia, Goiás, Brazil (16º45'45.2" S, 49º07'06.8" W and at an elevation of 762 m above sea level). Prof. Dr. José Realino de Paula identified the botanical material. A voucher specimen was deposited at the Herbarium of the Federal University of Goiás/Brazil under registration 60010. The leaves were air dried at room temperature for three days, triturated using commercial crusher (Skymsen, LS-08MB-N) immediately prior to the extractionof the volatile oil.

2.2 Extraction and analysis of volatiles oils by GC/MS

115g of the powdered material (leaves collected in the two months) was subjected to hydro-distillation for three hours in a Clevenger type apparatus. The VO obtained was dried with anhydrous Na2SO4 and kept at -18 °C until analysis. The successive extractions of VO were collected in a single sample and then analyzed by gas chromatography coupled to mass spectrometry (GC/MS) in a QP2010 Plus (Shimadzu Corporation, Japão), equipment with a linear quadrupole mass detector, DB-5MS chromatography column (30 m x 0.25 mm, 0.25 μm); 5% phenylmethylpolysiloxane (Agilent J & W GC columns, USA). Helium was used as carrier gas at a flow rate of 1ml min. The injector and interface temperatures were 225 °C and 240 °C, respectively, with a 1:20 split ratio. The injection volume was 1 μL sample in hexane (1:5) and the oven temperature program consisted of a ramp of 60-240 ºC/3 ºC/min; 240-280 °C/10 °C/min, and final temperature set at 280 °C for 8 min. The temperature of the ion source 240 °C and electron impact ionization, 70 eV; the acquired mass range was 40-350 m/z. The chemical constituents of VO were identified by comparison of its retention index (RI), as well as of its mass spectra described in the literature. Retention indices were obtained with reference to a linear sequence of C7-C30 n-alkanes (Sigma, USA) (Adams, 2007).

2.3 Preparation of emulsion

16 emulsions were prepared with VO, arabic gum (AG) (Sigma-Aldrich) and maltodextrin dextrose (MD) equivalent 4.0-7.0 (Sigma-Aldrich) under constant magnetic stirring until complete solubilization. The 16 emulsions were prepared in a proportion VO: AG: MD (w/w) (1:2.7:1.5). The ratio of volatile oil /wall material was 0.25, the solids content ranged 32.8 to 34.59% and the VO percentage ranged 7.75 to 9.14%. After incorporation of the wall material, the blend was homogenized in ULTRATURRAX, IKA® brand and digital T25 model, operating at 15.000 rpm for 10min and spray-drying was performed.

2.4 Spray-drying process

Spray drying was performed (Laboratory of Agricultural Engineering - CCET/UEG, Anápolis/GO) using spray-dryer LABMAQ model MSD 1.0, equipped with cylindrical camera of 50 cm of height by 9 cm of diameter; double fluid type conical sprinkler and feed system made by a peristaltic pump. The emulsions were atomized, dried and the microcapsules obtained were packed in an amber bottle at room temperature in a desiccator until further analysis.

2.5 Box–Behnken experimental design

The Box–Behnken experimental design was employed to optimize and evaluate main effects, interation effects and quadratic effects of the process variables on the quality of microcapsules. A design 33 was used to investigating quadratic response surfaces and obtaining second order polynomial models for optimization. The parameters investigated and their levels were: drying temperature (DT; 145-175 ºC), emulsion feed rate (FF; 0.25-0.35 L/h) and nozzle diameter (ND; 0.7-1.2 mm). Yield of the drying process (YP), volatile oil retention on the surface of the microcapsules, encapsulation efficiency (EE), moisture, water activity (Aw), apparently density, β-caryophyllene (CY) content and relative percentage of the major chemical compounds in VO recovered from microcapsules were evaluated as response variables.

A second-order polynomial model (Equation 1) was used to express the relationship between the variables investigated as a function of the independent variables, as follows:

$$Y_n = \beta_0 + \beta_1 X_1 + \beta_2 X_2 + \beta_3 X_3 + \beta_{12} X_1 X_2 + \beta_{13} X_1 X_3 + \beta_{23} X_2 X_3 + \beta_{11} X_1^2 + \beta_{22} X_2^2 + \beta_{33} X_3^2 \quad (1)$$

Where Y represents the response variables, β0 is an intercept, β1, β2 and β3 are linear coefficients, β11, β22, β33 are square coefficients, β12, β13, β23 represent interaction coefficients and X1, X2, X3 represent the levels of independent variables. This response surface model was also employed to predict the maximum encapsulation efficiency of the three-dimensional surface (3D), which is the projection of the response surface in a 3D plane (Wei, 1978; Derringer & Suich, 1980).

To optimize the spray-drying drying process, the desirability function method was used to find the optimum conditions of the analyzed responses (Derringer & Suich, 1980; Battista, et al., 2017).

The individual desirability functions (Equation 2) from the considered responses were combined to obtain the overall desirability D, defined as the geometric average of the individual desirability:

$$\mathbf{D} = (d1, d2, \ldots, dk)^{1/k} \quad (2)$$

The software Statistica 7 was used for analyses. Only the factors with $p < 0.05$ were considered.

2.6 Microcapsules characterization

2.6.1 Physico-chemical analysis

The water activity (Aw) of the microcapsules was performed by direct reading in a water activity analyzer of the AQUALAB brand, Water Activity Analyzer model. The apparently density (da) (g/cm) was determined by the test tube method according to.[40] The moisture content of the microcapsules was performed by the gravimetric method in scale with OHASUS MB-35 halogen lamp. In petri dish, 1.0g of the powder was heated to 105 ºC until constant weight. The percentage of moisture was calculated on a wet basis. All analyzes were performed in triplicate.

2.6.2 Encapsulation Efficiency

The EE was determined by the fraction of oil encapsulated on the amount of total oil (Equation 3) (Tan, et al., 2005).

$$EE(\%) = \frac{(VOT-VOS)}{VOT} x100 \quad (3)$$

Where EE is the encapsulation efficiency, VOT is the amount of total oil and VOS is the amount of surface (unencapsulated) oil on the surface of the microcapsules. The non-encapsulated VO present on the particle surface was determined with 5.0 g of microcapsules in a 125 ml capped flask and 20 ml of hexane added, the mixture stirred for 10 min at room temperature. The mixture was filtered on Whatman filter paper number 1 and the retained microcapsules were washed with 10 ml of hexane three times. The microcapsules were then oven dried at 50 ° C, weighed to a constant mass. The mass of unencapsulated VO was calculated by the difference in mass of the microcapsules before and after washing with hexane. The amount of total oil in the encapsulated products was determined by hydrodistillation of 5.0 g of the encapsulated powder for 3 h in Clevenger type apparatus. After extraction, the volume of VO was measured in the graduated tube of the apparatus itself. The total mass of the oil was calculated from the specific mass and volume (Tan, et al., 2005; Bae & Lee, 2008).

2.6.3 Volatile oil retention

The percent retention of VO in the microcapsules was determined in relation to the blend, prior to emulsification. The calculation was obtained by the ratio of the total oil quantified in the particles after drying by spray drying with the total oil initially added to the emulsion preparation, according to Equation 4 (Frascareli et al., 2012; Costa, et al., 2013).

$$RO(\%) = \frac{VOT}{VOE} x100 \quad (4)$$

2.6.4 Determination of the β-caryophyllene content in the VO of the of S. odoratissima leaves and of the microcapsules by HPLC

The quantification of β-caryophyllene in the VO of the microcapsules was performed according to the method developed by Alves, et al. (2014) using high performance liquid chromatography (HPLC-PDA) using acetonitrile (ACN), methanol (MeOH) and ultrapure water (Milli-Q® H2O) in a ratio of 75:10:15 (v/v) as the mobile phase at a flow of 1.3 ml/min at room temperature. Chromatography was performed using a C8 column (250 mm x 4.6 mm, 5 mm, Phenomenex®). The system was maintained under isocratic conditions. HPLC analysis was performed with a Waters® (Massachusetts, USA) chromatograph equipped with a separation module e2695 and a photodiode array detector (PDA 2998) defined at 210 nm. Empower 2.0 software was used for data collection and processing. In a 25 ml volumetric flask was added 0.1 g of the microcapsules and HPLC grade

methanol to the final volume. The mixture was subjected to ultrasonic extraction for 45 min. The solution was filtered through Whatman number 1 Filter Paper and Millex1 syringe filters (0.45 mm pore size) and then injected into the HPLC.

2.6.5 Morphological analysis of the microcapsules

For the morphological analysis, the microcapsule samples were fixed in metallic specimen holders (stubs) in carbon double-sided adhesive tape. It was then covered with a thin layer of gold metallic alloy in a Denton Vacuum sputter, Desck V and the time of gold deposition on the sample was 2 min., generating a layer of approximately 250 A. After metallization, the samples were observed on a JEOL® scanning electron microscope (JVM) model JSM-6610, equipped with EDS, Thermo scientific NSS Spectral Imaging. The analysis was performed at the High Resolution Microscopy Multiuser Laboratory (LabMic) at the Institute of Physics of the Federal University of Goiás (IF/UFG).

The particle size distribution analysis was performed using the ImageJ 1.50 ie software represented by the frequency of the intervals using Equation 5, where d is the diameter of the microcapsules (μm) and A is the area of the microcapsules (μm).

$$d = 2 (A/\pi)^{1/2} \quad (5)$$

3. Results

3.1 Volatiles oils by GC/MS

It was verified in VO from *S. odoratissima* leaves: 84.01% of sesquiterpene hydrocarbons, 11.35% of oxygenated sesquiterpenes and 0.13% of monoterpene hydrocarbons. The major compounds identified were bicyclogermacrene (15.53%), δ-cadinene (15.10%), β-caryophyllene (10.56%) and cis-muurola-4 (14), 5-diene (13.02%) (Table 1).

After the ME, there was a small change in the percentage of chemical compounds. In the recovered VO the sesquiterpene hydrocarbons ranged from 75.07 to 87.35%, with higher values in the samples of microcapsules obtained with lower drying temperature (145 °C). Oxygenated sesquiterpenes ranged from 9.96 to 15.32% and monoterpene hydrocarbons ranged from 0 to 0.17%. The main chemical components were β-caryophyllene (9.15-11.65%), bicyclogermacrene (14.58-21.21%), δ-cadinene (11.88-20.40%) and cis-muurola-4(14),5-diene (8.94-14.97%) (Table 1).

Table 1 - Percentage of chemical constituents of VO of *S. odoratissima* leaves and VO recovered from microcapsules, obtained by GC/MS.

Constituents	KI	VO in the plant (before encapsulating)	VO recovered from microcapsules (after encapsulation)															
			1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16
α-Pinene	939	0.13	-	0.08	0.09	-	0.08	0.07	0.07	-	-	0.09	0.08	-	0.09	-	-	-
Limonene	1029	-	-	0.06	0.08	-	0.08	-	0.07	-	-	0.07	0.04	-	-	-	-	-
δ-elemene	1338	1.95	2.65	0.97	2.06	2.48	2.08	1.84	1.84	2.65	2.67	2.16	1.89	1.85	1.76	2.49	2.42	1.78
α-cubebene	1348	0.67	0.65	0.75	0.88	0.83	0.66	0.79	0.71	0.72	0.71	0.51	0.50	0.52	0.63	0.71	0.65	0.80
α-ylangene	1375	-	-	-	0.14	-	0.09	-	0.09	-	-	0.07	-	-		-	0.11	
α-copaene	1376	2.85	2.74	0.99	3.50	3.40	2.59	3.34	2.91	2.98	2.87	2.05	2.19	2.43	2.68	2.86	2.76	3.36
β-bourbonene	1388	0.24	0.31	0.26	0.34	0.34	0.28	0.22	0.26	0.30	0.31	0.25	0.19	0.20	0.21	0.30	0.31	0.26
β-elemene	1390	3.33	4.30	3.68	3.67	4.14	3.63	3.31	3.15	4.18	4.14	3.55	3.53	3.30	3.03	3.98	4.03	3.32
β-caryophyllene	1419	10.56	9.15	10.61	10.18	9.97	11.03	11.21	11.40	9.89	10.24	10.59	11.41	11.42	10.43	10.57	10.45	11.65

β-copaene	1432	0.57	0.65	0.68	0.79	0.66	0.67	0.65	0.60	0.65	0.62	0.58	0.45	0.50	0.55	0.57	0.62	0.59
Aromadendrene	1441	0.8	1.04	0.98	1.35	1.05	1.11	0.91	1.04	0.94	0.92	0.91	0.77	0.80	0.81	0.89	0.84	0.89
cis-muurola-3,5-diene	1450	0.43	0.42	0.41	0.48	0.40	-	0.43	0.39	0.44	0.41	-	0.32	0.41	0.37	0.35	0.33	0.37
trans-muurola-3,5-diene	1453	0.53	0.52	0.36	-	0.49	-	0.39	-	0.54	0.49	-	0.27	0.33	0.31	0.43	0.41	0.34
α-humulene	1454	3.22	4.13	4.16	4.51	3.82	4.81	3.83	3.75	3.80	3.71	4.66	3.85	3.64	3.44	3.50	3.53	3.43
allo-aromadendrene	1460	2.85	3.72	3.48	3.51	3.75	3.21	3.33	2.93	3.77	3.61	2.88	2.88	3.08	3.03	3.39	3.28	3.24
cis-muurola-4(14),5-diene	1466	13.02	8.94	10.74	13.38	10.02	14.64	12.13	14.97	11.30	11.34	14.60	10.90	13.04	12.00	10.60	12.80	12.17
β- selinene	1490	0.65	0.96	1.11	-	0.85	-	0.96	-	0.84	0.85	-	0.89	0.85	0.86	0.78	-	0.83
Trans-muurola-4(14),5-diene	1493	1.84	1.33	-	-	1.43	-	-	-	1.46	1.29	-	-	-	-	1.18	-	-
Bicyclogermacrene	1500	15.53	14.58	18.07	18.70	15.64	20.07	19.27	21.21	16.32	16.66	20.48	19.81	18.59	17.85	17.08	18.10	17.75
α-muurolene	1500	2.74	-	-	-	-	-	-	-	-	-	-	-	-	-	0.20	-	-
β-trans-guaiene	1502	0.19	0.25	-	-	0.22	-	0.25	-	0.23	0.23	-	-	0.23	0.23	-	-	-
Germacrene A	1509	0.11	0.17	0.61	-	0.15	0.62	0.31	0.44	0.12	0.13	0.57	0.27	0.28	0.29	-	0.08	0.27
γ-cadinene	1513	5.05	5.12	-	5.63	5.31	-	4.88	-	5.25	5.07	-	4.43	-	4.83	5.16	-	-
δ-cadinene	1523	15.1	11.9	17.9	12.6	13.0	17.5	13.9	20.4	12.4	12.4	16.4	13.6	14.3	15.2	14.5	13.1	16.1
Zonarene	1529	0.5	-	-	-	-	-	0.58	-	0.69	0.73	-	-	0.52	0.60	-	-	-
trans-cadina-1,4-diene	1534	0.4	0.4	0.4	-	0.4	0.44	0.43	0.41	0.42	0.37	0.39	0.27	0.31	0.41	0.32	0.36	0.37
α-cadinene	1538	0.9	0.9	0.8	1.3	0.9	0.69	0.74	0.65	0.83	0.78	0.61	0.63	0.63	0.73	0.74	0.70	0.70
α-calacorene	1545	-	0.3	0.3	0.3	0.20	0.23	0.11	0.20	0.12	0.11	0.26	0.18	0.08	0.11	0.14	0.31	0.10
cis-muurol-5-en-4-β-ol	1551	-	-	-	0.08	-	0.06	-	0.05	-	-	0.06	-	-	-	-	-	-
Espatulenol	1578	1.42	2.87	2.39	1.88	2.23	1.79	1.61	1.44	1.84	1.97	1.76	2.34	1.58	1.78	2.14	-	1.49
Globulol	1590	0.24	0.83	0.48	0.51	0.51	0.44	0.36	0.36	0.42	0.54	0.62	0.81	0.59	0.43	0.41	0.59	0.29
Viridifloral	1592	-	-	-	0.47	-	0.38	-	0.31	-	0.27	0.50	-	0.34	-	-	-	-
Cubeban-11-ol	1595	-	-	-	-	-	-	0.08	-	0.09	-	-	-	-	-	-	-	-
Ledol	1602	0.14	0.34	0.25	-	0.29	-	0.21	-	0.18	-	-	0.27	-	0.22	0.25	0.28	0.18
1,10-di-epi-cubenol	1619	0.26	0.52	0.36	0.39	0.42	0.32	0.30	0.26	0.37	0.39	0.43	0.39	0.27	0.31	0.36	0.41	0.27
Junenol	1619	-	-	0.05	0.07	0.12	-	-	0.04	-	-	-	-	-	-	-	0.17	-
1-epi-cubenol	1628	0.45	0.79	0.62	0.63	0.63	0.61	0.41	0.46	0.58	0.60	0.83	0.62	0.42	0.50	0.56	0.62	0.41
α-epi-cadinol	1640	2.29	3.46	5.35	4.93	2.76	4.97	4.55	4.08	2.78	2.84	5.99	5.83	4.07	4.87	2.68	6.10	3.99
α-epi-muurolol	1642	1.86	5.50	4.38	4.16	4.83	4.10	3.84	3.43	4.89	4.77	5.13	4.99	3.93	4.11	4.59	5.30	3.33
α-muurolol(Torreiol)	1646	0.64	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
α-cadinol	1654	4.05	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Monoterpene hydrocarbons		0.1	0.0	0.1	0.2	0.0	0.2	0.1	0.1	0.0	0.0	0.16	0.12	0.0	0.1	0.0	0.0	0.0
Sesquiterpene hydrocarbons		84.0	75.1	77.3	83.3	79.4	84.4	83.9	87.4	80.9	80.7	81.6	79.2	77.3	80.4	80.8	75.1	78.3
Oxygenated sesquiterpenes		11.4	14.3	13.9	13.1	11.8	12.7	11.4	10.4	11.2	11.4	15.3	15.3	11.2	12.2	10.9	13.5	9.9
Total identified		95.5	89.4	91.3	96.6	91.2	97.2	95.3	97.9	92.0	92.0	97.0	94.6	88.5	92.7	91.8	88.6	88.3

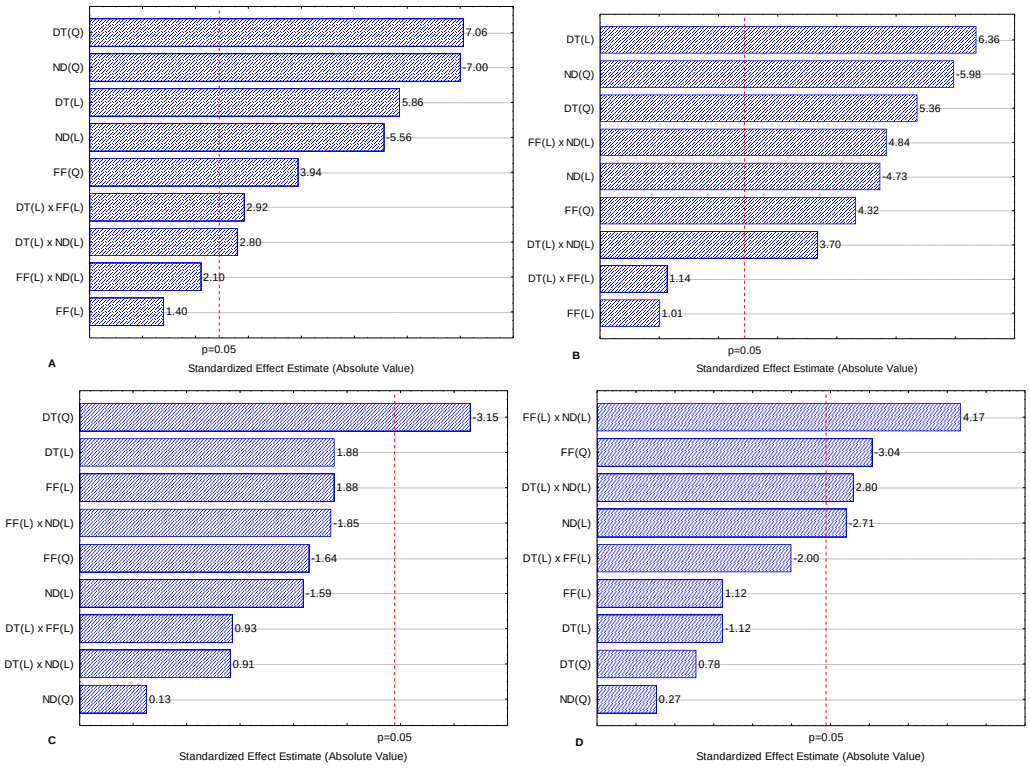
Legend: - (-) not identified; IK- Kovats index retention; VO- volatile oil. Source: Authors.

3.2 Statistical analysis of experimental design

The effects of the drying temperature, feed flow (FF) and nozzle diameter (ND) variables on the dry matter yield (YP), encapsulation efficiency (EE), retention of total volatile oil (RO), and β-caryophyllene content (CY) responses were tested for adequacy of analysis of variance (ANOVA).

The goodness of the model can be checked by the determination coefficient (R2). The R2 and adjusted R2 were 0.9500 and 0.8928 for dry matter yield; 0.9619 and 0.9183 for retention of VO; 0.8896 and 0.7634 for β-caryophyllene content, respectively, these values are indicative of a suitable model. The values of R2 indicates good relation between the experimental and predicted values of the response. The dry matter yield and retention of total volatile oil variables presented statistically significant effects at the 5% level in the microencapsulation process, for the conditions of drying temperature, drying process and feed flow (Figures 1A and 1B, respectively). However, EE showed a significant effect only for drying temperature (Figure 1C) and β-caryophyllene content showed a significant effect only on feed flow and drying process (Figure 1D).

Figure 1. Pareto chart of the standardized effects for the responses: (A) Drying process yield (YP); (B) Oil retention (RO); (C) Efficiency of encapsulation (EE) and (D) β-caryophyllene (CY) content.



Source: Authors.

For yield of the drying process (YP) (Equation 6) the drying temperature showed positive linear effect and negative quadratic effect. Similar behavior occurred with feed flow (FF), with positive linear effect and negative quadratic effect. The drying process nozzle diameter had a negative linear effect and a positive quadratic effect, and there was no statistically significant relationship between feed flow and nozzle diameter. For retention of total VO (Equation 7) the drying temperature and feed flow variables presented positive linear effects and a quadratic negative effect. The drying process variable had a linear negative effect and a positive quadratic effect. There was no statistically significant relationship between drying temperature and FF. In the β-caryophyllene content (Equation 8), the drying temperature variable had linear and quadratic negative effects, while feed flow had its positive effects. The nozzle diameter had negative linear effect but did not present a quadratic effect.

$$YP = 22.17 + 2.82DT + 0.68\ FF - 2.68ND + 1.99DTFF + 1.91DTND - 4.81DT^2 - 2.68FF^2 + 4.77ND^2 \quad (6)$$

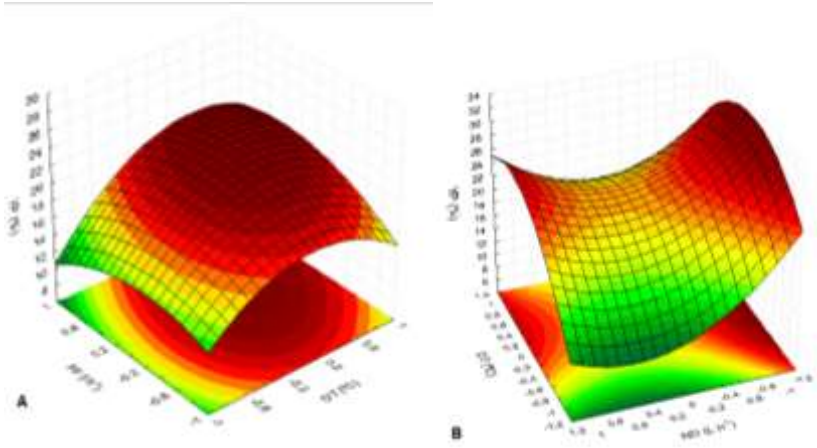
$$RO = 47.33 + 6.72DT + 1.07FF - 5.00ND + 5.50DTND + 7.24FFND - 8.01DT^2 - 6.46FF^2 + 8.94ND^2 \quad (7)$$

$$CY = 6.35 - 0.28DT + 0.28FF - 0.69ND - 0.72DTFF + 1.01DTND + 1.50FFND - 0.28DT^2 + 1.09FF^2 \quad (8)$$

3.3 Scanning Electron Microscopy (SEM)

The effects of the independent variables drying temperature, feed flow (FF), nozzle diameter (ND) and their interactions on the dry matter yield (YP), retention of total VO (RO) and β-caryophyllene content (CY) responses can be visualized on the surface response graphs. In relation drying temperature and feed flow (Figure 2A) the best dry matter yield was found in the central point. In the relationship between drying temperature and nozzle diameter (Figure 2B) the best dry matter yield was obtained with lower values of nozzle diameter over a wide temperature range and with higher nozzle diameter and higher temperatures. There was no statistical relationship between feed flow and nozzle diameter.

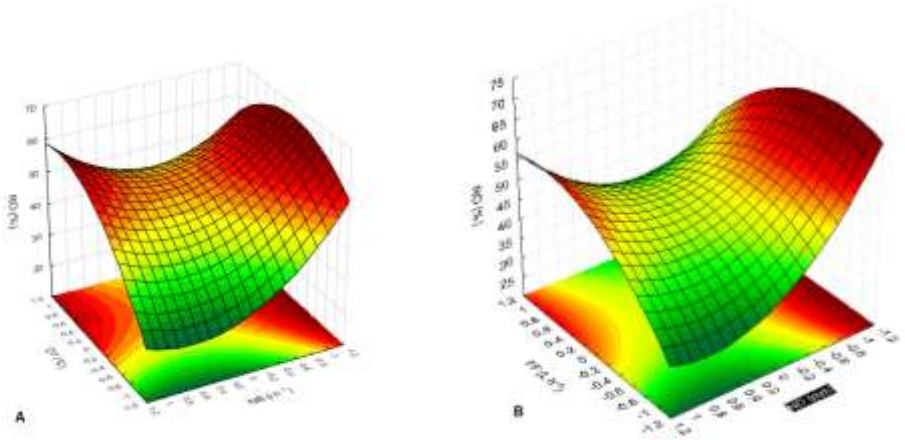
Figure 2. Response surface generated by the quadratic mathematical model for the drying process Yield (YP) by spray-drying of the volatile oil of *S. odoratissima*. Optimization of the variables: (A) temperature and feed flow (FF), (B) temperature and diameter of the spray nozzle (ND).



Source: Authors.

The increase in retention of total VO was observed for smaller values of nozzle diameter in a wide range of the drying temperature domain, with emphasis on the region of the graph (Figure 3A) with intermediate temperature, obtaining microcapsules with 61.57% retention of total VO (sample 9, Table 2). However, the association of higher values of drying temperature and nozzle diameter produced microcapsules with a high retention of total VO of 55.02% (sample 8, Table 2). For the ratio of feed flow and nozzle diameter (Figure 3B), the variable that had a significant effect on the retention of total VO was the nozzle diameter, with lower levels of nozzle diameter associated with intermediate feed flow values, we obtained rates highest retention of total VO of 52.49% (sample 5, Table 2). No significant interaction was observed between the feed flow and the nozzle diameter.

Figure 3. Response surface for the VO retention of *S. odoratissima* (RO) by spray-drying. Optimization of the variables: (A) temperature drying temperature (DT) and spray nozzle diameter (ND). (B) feed flow (FF) and nozzle diameter (ND).



Source: Authors.

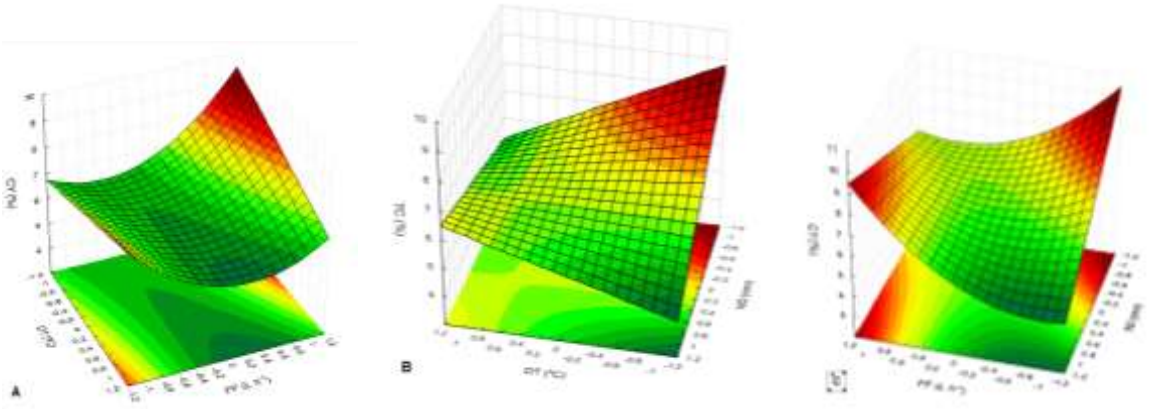
Table 2. Physical-chemical characterization of the VO microcapsules of *S. odoratissima* leaves obtained by spray-drying.

Experiment	EE (%)	Moisture (%)	A_w	d_a (g cm^{-3})	CY (%)
1	77.75±0.43	1.97±0.12	0.28±0.02	0.53±0.03	6.48±0.23
2	76.43±0.49	1.74±0.22	0.23±0.02	0.48±0.01	6.99±0.53
3	74.8±1.24	2.04±0.04	0.25±0.02	0.56±0.01	8.86±0.49
4	80.81±3.76	3.04±0.18	0.21±0.01	0.49±0.03	6.5±0.35
5	75.34±3.25	3.26±0.26	0.25±0.01	0.55±0.04	8.07±0.36
6	79.89±4.44	3.03±0.32	0.22±0.01	0.50±0.02	5.85±0.25
7	64.43±1.82	3.69±0.18	0.28±0.02	0.59±0.01	4.18±0.47
8	76.16±2.49	3.58±0.19	0.22±0.01	0.47±0.03	5.98±0.27
9	63.24±12.73	4.40±0.28	0.22±0.05	0.50±0.03	9.23±0.63
10	80.28±2.44	2.25±0.54	0.17±0.03	0.50±0.01	6.42±0.15
11	68.98±1.53	3.77±0.15	0.24±0.02	0.53±0.04	5.36±0.32
12	71.44±2.18	3.86±0.26	0.22±0.02	0.55±0.04	8.55±0.33
13	69.43±0.54	3.52±0.14	0.22±0.01	0.57±0.04	6.88±0.29
14	70.79±0.92	3.36±0.17	0.22±0.01	0.58±0.04	6.9±0.64
15	65.95±4.19	2.34±0.18	0.21±0.08	0.54±0.01	5.17±0.17
16	65.83±2.45	3.29±0.16	0.24±0.02	0.46±0.04	6.63±0.29

Legend: EE-encapsulation efficiency; A_w -water activity; d_a-apparent density; CY-β-caryophyllene content by HPLC. Source: Authors.

In the interaction of drying temperature and feed flow (Figure 4A), the highest levels of β-karyophylene were observed for lower values of drying temperature and higher values of feed flow. In the interaction between drying temperature and nozzle diameter (Figure 4B), the lower levels of drying temperature and nozzle diameter resulted in higher values of β-karyophylene content and in the interaction between the feed flow and the nozzle diameter, at the lower levels of these two factors, the higher the value of β-karyophylene content (Figure 4C).

Figure 4. Response surface for the spray-drying β-caryophyllene content of the volatile oil of *S. odoratissima*. Optimization of the variables: (A) drying temperature (DT) and feed flow (FF). (B) temperature (DT) and nozzle diameter (ND). (C) feed flow (FF) and spray nozzle diameter (ND).



Source: Authors.

3.4 Optimization of microencapsulation

The optimal conditions of the drying process for the VO microencapsulation of *S. odoratissima* leaves were obtained based on the Desirable function D represented best solutions for the system of the dry matter yield, retention of total VO and β-caryophyllene content responses (Table 3): DT (158 °C), feed flow (0.25 L/h) and drying process (0.7 mm).

Tabela 3. Box–Behnken design and observed responses to the microcapsules of the *S. odoratissima* leaves.

Experiment	DT (ºC)	FF (l h^{-1})	ND (mm)	Responses		
				YP (%)	RO (%)	CY (%)
1	145.0	0.25	1.0	12.76	25.73	6.48±0.23
2	175.0	0.25	1.0	14.68	38.30	6.99±0.53
3	145.0	0.35	1.0	10.69	24.01	8.86±0.49
4	175.0	0.35	1.0	20.58	43.38	6.50±0.35
5	145.0	0.30	0.7	23.69	52.49	8.07±0.36
6	175.0	0.30	0.7	25.27	52.40	5.85±0.25
7	145.0	0.30	1.2	15.19	33.11	4.18±0.47
8	175.0	0.30	1.2	24.39	55.02	5.98±0.27
9	160.0	0.25	0.7	28.32	61.57	9.23±0.63
10	160.0	0.35	0.7	26.24	49.68	6.42±0.15
11	160.0	0.25	1.2	19.42	35.46	5.36±0.32
12	160.0	0.35	1.2	23.08	52.52	8.55±0.33
13	160.0	0.30	1.0	21.34	46.88	6.88±0.29
14	160.0	0.30	1.0	22.14	51.70	6.90±0.64
15	160.0	0.30	1.0	24.66	47.39	5.17±0.17
16	160.0	0.30	1.0	20.56	43.35	6.63±0.29

Legend: DT-inlet temperature; FF-feed flow; ND-atomizer nozzle diameter; YP-yield of the drying process; RO-retention of volatile oil; CY-β-caryophyllene content by HPLC. Source: Authors.

3.5 Experimental verification

The verification experiments (in triplicate) had been performed for the optimum point mathematically determined in the optimization process (D = 0.96): dry matter yield (20.3±0.23%), retention of total VO (31.27±3.75%) and β-caryophyllene content (7.46±0.31%). Thus, the second order models were adequate to describe the influence of the operational variables employed in the mathematical model. The others parameters analyzed were disregarded because they were out of point mathematically determined in the optimization process: EE (64.03±10.14), humidity (4.01±0.70), Aw (0.15±0.01) and da (0.23±0.03).

3.6 Physico-chemical of microcapsules

In the triplicate assay by HPLC, the β-caryophyllene content (CY) in the VO of *S. odoratissima* leaves was 14.57 ± 0.89% and the β-caryophyllene content recovered from the microcapsule ranged from 4.18±0.47 to 9.23±0.63% (Tables 2 and 4). In the physicochemical characterization of the microcapsules, the results found varied from 0.17±0.03 to 0.28±0.02; 0.4±0.04 to 0.59±0.0 g/cm; and 1.74±0.22 to 4.40±0.28% for the analysis of water activity (Aw), bulk density and moisture content, respectively. Regarding encapsulation efficiency (EE) in the ME, the values ranged from 63.24± 2.73 to 80.81±3.76%. Samples 10 and 4 presented the best results for EE, higher than 80% (Table 4) with feed flow (FF) of 0.35 L/h and drying temperature of 160 and 175 °C, respectively (Table 2). All experiments were performed in triplicate.

Table 4. Optimized spray-drying drying conditions and physicochemical analyzes of microcapsules.

Drying parameters	DT (ºC)	158
	FF (l h^{-1})	0.25
	ND (mm)	0.7
Responses	YP (%)	20.31±0.23
	RO (%)	31.27±3.75
	CY (%)	7.46±0.31

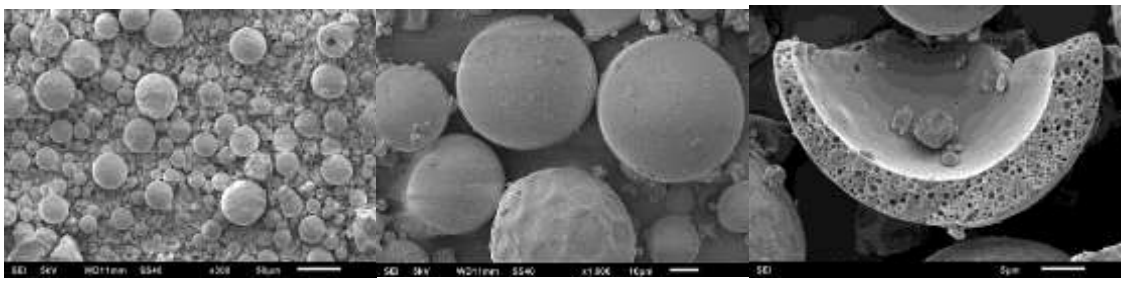
Legend: DT- drying temperature; FF- feed flow; ND - atomizer nozzle diameter; YP - yield of the drying process; RO - retention of volatile oil; CY - β-caryophyllene content. Source: Authors.

3.7 Morphological of the microcapsules

The scanning electron microscopy (SEM) (Figure 5) shows the microcapsules obtained under the optimized conditions of drying by spray-drying, revealing spheres with small vacuoles, little roughness on the surface, homogeneous appearance, no cracks or breaks and thick walls with small pores. No deformations and microcapsules adhered to each other were observed.

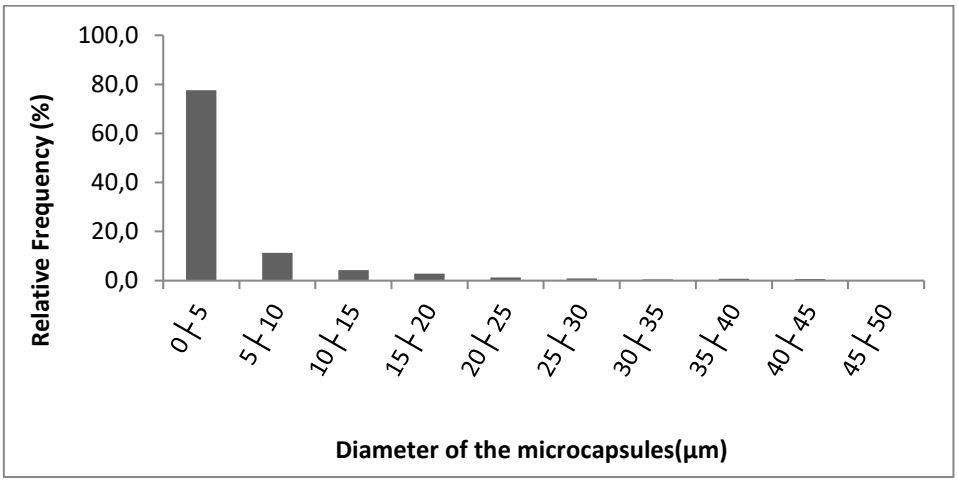
The distribution of the microcapsule diameter (Figure 6) of VO of *S. odoratissima* leaves showed variation from 0.65 to 50.34 μm for a sample field of 2134 particles, and 77.6% of them are contained in the shortest range of size with a diameter varying from 0-5 μm.

Figure 5. Scanning Electron Microscopy of the volatile oil microcapsules from *S. odoratissima* leaves in the optimized conditions: A) field image of microcapsules; B) microcapsule surface and C) microcapsule wall.



Source: Authors.

Figure 6. Graphical representation of the relative frequency distributions obtained from 2134 particles observed in the set of images composed of volatile oil microcapsules from *S. odoratissima* leaves.



Source: Authors.

4. Discussion

The use of gum arabic (GA) and maltodextrin (MD) as a wall material in *S. odoratissima* VO microencapsulation produced a white powder characteristic of microencapsulated product. Other studies on ME of VO by spray-drying produced microcapsules with similar appearance as for *Backhousia citriodora* Hook & Harv. (Bringas, et al., 2012), *Pterodon emarginatus* Vogel (Alves, et al., 2014), *Cinnamomum zeylanicum* (Felix, et al., 2017) and *Rosmarinus officinalis* (Fernandes, et al., 2014).

The chemical composition of the OV from *S. odoratissima* leaves presented four major compounds, bicyclogermacrene (15.53%), δ-cadinene (15.10%), β-caryophyllene (10.56%) and cis-muurola-4 (14), 5-diene (13.02%). These compounds were selected to monitor the chemical profile of OV and OV recovered from dry microcapsules by spray drying at concentrations higher than 10%. The lowest percentages of bicyclogermacrene (14.58%), δ-cadinene (11.88%), β-caryophyllene (9,15%) and cis-muurola-4 (14), 5-diene (8,94%) of volatiles in ME were observed in sample 1, where the drying parameters employed were atomizer nozzle diameter (1.0mm) and lower values for drying temperature lower drying temperature (145ºC) and lower feed flow (0.25 L/h). Even with losses, this combination ensured balance between the rate of evaporation of the water and the slow formation of outer film and therefore no crust formation with fissures (Frascareli, et al., 2012; Gonçalves, et al., 2016). Alves, et al. (2014) obtained loss of 35.74%, 39.50%, 29.95% and 44.71% β-caryophyllene in *Pterodon emarginatus* VO microcapsules using the same wall material (AG and MD) under conditions of drying temperature drying (130ºC), nozzle diameter (1.2mm) and feed flow (4mL/ min), respectively.

The α-muurolol and α-cadinol compounds were identified only in the VO of *S. odoratissima*. These differences can be attributed to the spray-drying process (Asensio, et al., 2017), suggesting that the drying parameters applied to the ME of volatile oil of *S. odoratissima* leaves caused loss by volatilization, thermal degradation of these constituents or formation of unidentified artifacts (Alves, et al., 2014). According to Gharsallaoui, et al. (2007) and Ribeiro, et al. (2015), the loss of volatiles during ME, occurs predominantly in the initial drying stage.

In the present work a previous screening was done in search of the best proportions of wall material (GA:MD), wall and core material (WM: VO), VO percentage and solids content. The microcapsules that presented the best results for the encapsulation efficiency, retention of total volatile oil, dry matter yield, A_w, particle size distribution and morphological aspect were selected for the experimental design in the drying process.

For microencapsulation, a mixture of gum arabic (GA) and maltodextrin (MD) was selected because of the pleasant taste, low cost, besides being a polymer matrix that produces microcapsules with high encapsulation efficiency (Bora, et al., 2018; Mahdavi, et al., 2016).

Studies have shown the importance of drying factors to obtain microcapsules with physicochemical, chemical and morphological properties (Singh & Van, 2016) within quality standards for use in the food and pharmaceutical industries. The ranges of factors selected for experimental design (temperature, feed flow and spray nozzle diameter), were based on preliminary investigations in the literature, considering studies with VO of *Copaifera multijuga* (Dias, et al., 2012) and *Cinnamomum zeylanicum* (Felix, et al., 2017; Hermanto, et al., 2016), *Cymbopogon citratus* (Ribeiro, et al., 2015).

Following completion of the experiment, it was found that the water activity (A_w) in the microcapsules ranged from 0.17±0.03 to 0.28±0.02. The value of A_w <0.3 is ideal for stability, as this low amount of water on the surface of the microcapsules becomes unavailable and consequently increases the shelf life of the product. Low A_w values are a prerequisite for the commercial production of powders with good characteristics such as high flow capacity, low viscosity and particle agglomeration (Behboudi, et al., 2013)**.** Alves, et al. (2014) obtained VO microcapsules from fruits of *Pterodon emarginatus* with A_w values close to 0.2. Other works produced microcapsules with different encapsulating materials by spray-drying and obtained similar A_w values of the *S. odoratissima* microcapsules. Among them, studies with probiotic microcapsules (A_w = 0.11) (Behboudi, et al., 2013) and orange oil (A_w = 0.187) (Guzmán, et al., 2013). Daza, et al. (2016) verified in microcapsules of extract of *Eugenia dysenterica* DC. (Myrtaceae) under different drying conditions, A_w varying in the range of 0.12 to 0.26.

The 16 microcapsules samples had a range of 0.46±0.01 to 0.59±0.06 g cm^{-3}. These values were are higher to other studies that used drying ranges and similar encapsulating materials. Microcapsules obtained from linseed oil had a range of 0.28-0.40 g/ mL (Carneiro, et al., 2013); babassu milk of 0.390±0.003g/mL (Santana, et al., 2013); *Rosmarinus officinalis* L. volatile oil (Lamiaceae) (0.41–0.52g/mL) (Victória, et al., 2013) and resin oil of *Nigella sativa* L. (Ranunculaceaae) of 0.5±0.01g/mL (Edris, et al., 2016). The microcapsules obtained using the lower drying temperature factors (145ºC) and higher nozzle diameter (1.2mm) produced microcapsules with greater guarantee of less oxidation and consequent increase of the stability of the microcapsules, besides reducing costs of storage, transportation and commercialization of the product (Bakry, et al., 2016; Santana, et al., 2013).

The moisture content for VO the *S. odoratissima* microcapsules varied from 1.74±0.22 to 4.40±0.28%. Close values were found in VO microcapsules from *Rosmarinus officinalis*, with levels varying from 0.26 to 3.16% (Fernandes, et al., 2014) and 1.10 a 3.56% (Fernandes, et al., 2016). The moisture contained in the microcapsules represents low efficiency in the removal of solvents during drying and is directly related to DT (Oliveira & Petrovick, 2010). The microcapsules with lower moisture content (1.74%) were obtained with higher drying temperature (175ºC) and justified by the greater heat transfer to the particle, favoring the evaporation of water (Daza, et al., 2016). There was higher moisture content in the microcapsules when the feed

flow was higher. This occurred because the contact time of the emulsion with the drying air was not sufficient to cause evaporation of the water by virtue of the high amount of product to be dried (Fernandes, et al., 2014).

The encapsulation efficiency ranged from 63.24±12.73 to 80.21±3.76%, considering that of these values, superficial VO of the microcapsules was discarded. The VO present in the microcapsule indicates the efficiency of the spray-drying encapsulation process (Fernandes, et al., 2016; Martins, et al., 2014). The surface VO oxidizes and may compromise the quality of microcapsules (Edris, et al., 2016). The encapsulation efficiency of sample 4 (80.21%) of *S. odoratissima* is in accordance with other studies such as in the microencapsulation of coffee oil (82.57%) (Frascareli, et al., 2012), *Zingiber officinale* Rosco oil (31.19%) (Fernandes, et al., 2016) and *Juglans regia* L. oil (91.01%) (Shamaei, et al., 2017). Edris, et al. (2016) obtained 96.2% EE in *Nigella sativa* resin oil with 3.8% oil on the surface of the microcapsules.

After statistical evaluation, only dry matter yield, retention of total volatile oil and β-caryophyllene content presented significant responses as parameters to evaluate the quality of the microcapsules. This is relevant for phytopharmaceutical technology, which is challenging to produce microcapsules with desired content of available active compounds.

The β-caryophyllene content (CY) obtained in the VO of *S. odoratissima* leaves was 14.57±0.89% and the CY of the VO recovered from the microcapsules ranged from 4.18±0.47 to 9.23±0.63%. These values present index of degradation of the chemical marker varying from 36.65 to 71.31%. In the drying process, the volatility or degradation of some heat-sensitive chemical components may occur (Bakry, et al., 2016).

The yield of dry matter expresses the amount of powder recovered spray-dry by cyclone separation (Behboudi, et al., 2013). Both the feed flow and drying temperature impacted the recovery of microcapsules in the cyclone. The dry matter yield was maximized when the spray-dry was operated at intermediate temperature (160°C) and lower feed flow (0.25 L/h).

The higher concentration of AG in relation to MD and the solids content of the emulsion of 35% contributed to the good morphological appearance and encapsulation efficiency, producing smooth surface microcapsules with few cavities (depressions). According to Koç, et al. (2015) the vitreous properties of MD oppose the plastic properties of GA and together form spherical structures of smooth surfaces. Like the encapsulating material, feed flow can also affect the morphology of the microcapsules along with the airflow. These balanced factors guarantee the effective drying with elimination of the water, avoiding the roughness of the surface of the microcapsule.

The formation of rough surfaces in the microcapsules, according to Reineccius (2004) and Santana, et al. (2016) may occur depending on the drying rate and the cooling process of the microcapsules after swelling with the water vapor outlet.

In the present study, microcapsules with eventual breakage of the walls with an empty center (nucleus) were found, where the VO was trapped. According to Bakry, et al. (2016) and Martins, et al. (2014) this is a typical microcapsule structure of the reservoir type system. In addition, the small vacuoles on the walls suggest that droplets of the volatile oil were also trapped in the walls of the particles (Ramos, 2006).

In the drying process there is strong interaction between the parameters used in the microencapsulation. The size of the microcapsules is determined by the interaction of VO concentration, droplet size, drying temperature and feed flow (Singh & Van, 2016). A study by Littringer, et al. (2012) showed a decrease in microparticle size when an increase in flow rate was employed. The distribution of the VO microcapsule diameter of *S. odoratissima* leaves in the present study showed a higher percentage contained in the range of 0-3 μm (77.6%). According to Alves, et al. (2014), to be considered microcapsule, the particles in the form of powder should have diameters less than 30 μm. The size of the microcapsules is important to aid in their application as they may interfere with the texture, flavor and stability of the active principle (Comunian & Favaro, 2016), appearance and fluidity (Alves, et al., 2014; Fernandes, et al., 2014).

In the response surface methodology, the predicted model was adequate to describe the experimental domain and the reliability of the adjusted model. The optimum conditions found simultaneously for the response variables from the global Desirability function (D = 0.96) were: 20.31±0.23 for dry matter yield, 31.27±3.75 for RO and 7.46±0.31 for β-caryophyllene content, when drying temperature (158ºC), feed flow (0,25 L h^{-1}) and nozzle diameter (0,7mm) were used.

5. Conclusion

In conclusion, the best operational conditions for the atomization drying of *S. odoratissima* volatile oil were inlet temperature of 158°C, feed flow of 0.25L/h and drying nozzle diameter of 0.7mm. The Box-Behnken experimental model was able to identify the best operational conditions for the atomization drying of the volatile oil of *S. odoratissima*, guaranteeing good dry matter yield, retention of total volatile oil and β-caryophyllene. These results reveal the technological potential of the microcapsules obtained from *S. odoratissima* volatile oils. In addition, the process developed adds value to the raw material studied. For future studies, it is suggested to test the analgesic, anti-inflammatory, and antimicrobial activities from the microcapsules with the volatile oil of *S. odoratíssima* in order to analyze whether there is potentiation of their actions.

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