

Article - Health Science/Pharmacognosy

Lemongrass Essential Oil (*Cymbopogon citratus* (DC) Stapf.) Seasonal Evaluation and Microencapsulation by Spray-Drying

Thais Leticia Moreira da Silva¹ https://orcid.org/0000-0003-0001-2602

Gabriela latczaki da Rosa² https://orcid.org/0000-0002-9591-1418

Milena Aparecida Lorang dos Santos² https://orcid.org/0000-0002-2917-3867

Sara Lenz Graf³ https://orcid.org/0000-0003-4741-5783 Beatriz Helena Lameiro de Noronha Sales Maia³ https://orcid.org/0000-0001-5896-2892

Flávio Luís Beltrame^{1,2} https://orcid.org/0000-0001-7067-5802

Priscileila Colerato Ferrari^{1,2*} https://orcid.org/0000-0002-4285-6646

¹Universidade Estadual de Ponta Grossa, Programa de Pós-Graduação em Ciências Farmacêuticas, Ponta Grossa, Paraná, Brasil; ²Universidade Estadual de Ponta Grossa, Departamento de Ciências Farmacêuticas, Ponta Grossa, Paraná, Brasil; ³Universidade Federal do Paraná, Programa de Pós-Graduação em Química, Curitiba, Paraná, Brasil.

Editor-in-Chief: Paulo Vitor Farago Associate Editor: Jane Manfron Budel

Received: 06-Jan-2023; Accepted: 26-Jun-2023

*Correspondence: priscileila@hotmail.com; pcferrari@uepg.br; Tel.: +55-42-991310524 (P.C.F.)

HIGHLIGHTS

- High concentration of citral in pure and microencapsulated lemongrass essential oil.
- Warmer seasons (spring and summer) presented highest concentration of citral.
- Physicochemical evaluation of microcapsules containing lemongrass essential oil.
- Lemongrass essential oil microcapsulation by spray drying.

Abstract: Lemongrass (*Cymbopogon citratus*) is an aromatic plant of great significance in industries due to its essential oil characteristic aroma. Essential oils are concentrated plant extracts that evaporate when in contact with air and have low chemical stability, which can improve encapsulation techniques. This study aimed to evaluate the seasonal influence in the concentration of the predominant component citral of the essential oil of *C. citratus* and develop microparticles by spray drying containing this vegetal matrix. The lemongrass leaves were collected in the four seasons, and the essential oil was extracted by hydrodistillation. The chemical composition was determined by gas chromatography coupled with the mass spectrometry technique (GC-MS). Different proportions of arabic and guar gums were evaluated as encapsulating agents. Physicochemical analyses characterized the microparticle powder. GC-MS showed higher citral in the spring and summer (84.89% and 79.39%, respectively), and it was possible to identify a high amount of citral in the microparticles. Field-emission scanning electron microscopy showed agglomerate and collapsed microparticles. Fourier transform infrared spectroscopy suggested the essential oil encapsulation due to a

band at 1675 cm⁻¹ used as a reference, related to the acyclic monoterpenes of citral. X-ray diffraction showed the amorphous structure of the microparticles. Thermogravimetric curves of microparticles showed higher T_{onset} in microparticles than those observed for pure lemongrass essential oil, inferring that microencapsulation improved the thermal stability. This result confirms the lemongrass essential oil microencapsulation and its potential application in food products, drugs, or cosmetics.

Keywords: technological innovation; citral; arabic gum; guar gum; seasonal evaluation; gas chromatography-mass spectrometry.

INTRODUCTION

Plants have been used for a long time by industries and in academic research because they produce a wide and diverse variety of organic compounds through their metabolism. Especially the secondary metabolites, which do not participate directly in plant growth and development, are produced in a particular way by each species with various functions such as protection and defense against chemical, physical or biological agents [1]. Several environmental factors affect the production of these metabolites, such as seasonality, circadian rhythm, temperature, water availability, ultraviolet radiation, soil nutrients, altitude, and atmospheric composition [2]. Essential oils are secondary metabolites, most often obtained by hydrodistillation, composed of lipophilic and highly volatile substances. They included monoterpenes, sesquiterpenes, phenylpropanoids, and other low molecular weight substances, whose mixture confers the characteristic aroma of each plant [3].

Cymbopogon citratus (DC) Stapf., popularly known as lemongrass, belongs to the Poaceae family and is an example of an aromatic plant that produces essential oil. It originates from southwest Asia, but nowadays, it grows spontaneously worldwide, especially in tropical and subtropical regions [4]. They are used in ethnopharmacology for treating gastrointestinal and nervous disturbances and as an anti-inflammatory, analgesic, sedative, and antipyretic [5]. The lemongrass essential oil is extracted from dried leaves, with a yield of 0.5 to 2.0%, and the oil has at least 60% of citral, which consists of a mixture of two isomers: neral (Z-isomer) and geranial (E-isomer). Citral is the main component and the chemical marker of the *C. citratus* essential oil, and its concentration depends on geographical factors, which is higher in tropical regions [6,7].

Essential oils applications have been increased due to comprehensive requirements for pure natural ingredients, and they exhibit a wide range of biological activities, representing an actual potential product for the pharmaceutical industries. Also, it can be used as preserving and flavoring agents, as fragrance, and as aromatherapeutic agents in the food and cosmetic industries [8,9]. However, essential oils are challenging to manipulate and apply in the industry due to their low solubility in water, high volatility, and easy degradation by heat, oxidation, and light [10].

In order to increase the stability and protection of the essential oil from environmental agents, microencapsulation methods are used as an efficient technique and strategy for their commercial application possibilities. The spray-drying technique is one of the oldest encapsulation methods used to prepare the first flavor compounds encapsulated. This method is the most used in the food industry by presenting low costs of manufacturing, varieties of encapsulating materials, adequate retention and stability of volatile compounds, and large-scale production in a continuous mode [10]. This technique can produce microcapsules (diameter between 0.2 to 5000 μ m) through the atomization of tiny droplets of dispersion in spray-dryer equipment, which dries the solvent using hot air and solidifies, developing spherical structures with the active compound in the core, surrounded by a coating called wall material [11,12].

The encapsulating agent choice is critical in the spray-drying technique because it determines the physicochemical and morphological properties of the microcapsules. Different materials have been used for microencapsulation, especially biopolymers such as starches, cellulose derivatives, gums, gelatins, and lipids [13]. Among the numerous options, arabic gum is an encapsulating material used for microencapsulation by spray-drying due to its low viscosity and emulsifying properties [11]. However, it is common to mix wall materials to improve the process, and guar gum can also be used to form hydrogen bonds with water, increasing the solubility of lipophilic compounds and allowing a low cost [14]. Also, the spray-drying technique uses temperature in the process, which can degrade the components of the formulation. Therefore, maltodextrin, a biopolymer produced from starch, can be applied as a drying aid, protecting the core from temperature [15].

Thus, this work aimed to evaluate the seasonal influence in the yield and chemical composition of the lemongrass essential oil to determine the best season for extraction and to perform its microencapsulation by spray-drying technique, using arabic gum and guar gum as encapsulant agents and maltodextrin as a cryoprotective agent.

MATERIAL AND METHODS

Seasonal evaluation of lemongrass essential oil

The *C. citratus* leaves were collected in Ponta Grossa, Parana, Brazil (Latitude: $25^{\circ}04'05.0"$ S; Longitude: $50^{\circ}09'33.9"$ W), in the four seasons: winter (August/2021), spring (November/2021), summer (January/2022), and autumn (April/2022). A voucher specimen was deposited at the herbarium of the State University of Ponta Grossa, Parana, Brazil. The leaves were cut with scissors close to the stem and then spread on kraft paper to dry at room temperature ($\pm 22 \$ °C) for seven days. The dry leaves were weighed and cut to a length of approximately 2 cm. The essential oil extraction was performed by hydrodistillation process using distilled water in a Clevenger apparatus for 2 hours. The essential oil was collected from the apparatus and stored in a freezer (-11 °C) until its use. The following equation was used to calculate the yield of the process.

$$Y_{EO} = M_{EO}/M_{DL} \times 100 \tag{1}$$

 Y_{EO} corresponds to essential oil (EO) yield (% m/m), M_{EO} is the extracted essential oil mass, and M_{DL} is the dry leaves mass used in the extraction.

The essential oils' chemical composition was determined by a Shimadzu GC-MS chromatograph (Model GC-2010 Plus) coupled to a triple quadrupole tandem mass detector (Model TQ8040) and automatic injector (Model AOC-5000 Plus). The lemongrass essential oil was diluted to 1% (v/v) in hexane and analyzed using the following conditions: SLB-5MS fused silica capillary column (5% diphenyl + 95% dimethylpolysiloxane (30 m x 0.25 mm x 0.25 µm); the carrier gas used was helium with a flow rate of 1.02 mL.min⁻¹, with a split ratio of 1:90, with the injector at 250 °C and the ionization system at 70 eV. The sample was injected (1 µL) in the following ramp of heating: initial temperature 60 °C (0') to 250°C, with a heating rate of 3 °C.min⁻¹. A homologous series of saturated linear hydrocarbons, C8 to C19, was used to calculate the arithmetic index. The essential oils were identified by mass spectra and arithmetical indices comparison to the Gas Chromatography-Mass Spectrometry (GC-MS) library (NIST 14) and literature [16].

Microencapsulation of lemongrass essential oil by spray-drying technique

Arabic and guar gums dispersions in a hydroalcoholic solution (20% ethanol, v/v) were prepared in different proportions: 1:1, 3:1, and 1:3 (w/w). Tween[®] 80 (0.5%, w/v) was added as an emulsifier. Lemongrass essential oil (77.4% of citral; Quinarí[®], Brazil) in a 3:1 (w/v) ratio was added to the polymeric dispersion by mixing in a magnetic stirrer. Maltodextrin (6%, w/v) was added only before drying. Three microparticle formulations were prepared as control, composed of only one gum (arabic or guar gum) and a 1:1 arabic: guar gum (w/w) without the essential oil. The sample compositions are described in Table 1.

The samples were dried using a spray-dryer (LabMaq[®], model LM-MSD 0.5) under the following operational conditions: drying temperature of 80 °C, feed rate of 0.34 L/h, and an atomization pressure of 1.68 atm. The formulations were under constant agitation in a magnetic stirrer. The dried materials were collected from the cyclone and the drying chamber walls and stored in amber vials in the refrigerator (4 °C).

	lion		
Sample	Arabic/ guar gum	Lemongrass essential oil (mL)	Tween 80 (%)
AG/GG1:1-EO	1:1	10	0.5
AG/GG1:3-EO	1:3	10	0.5
AG/GG3:1-EO	3:1	10	0.5
AG-EO	1:0	10	0.5
GG-EO	0:1	10	0.5
AG/GG1:1	1:1	-	0.5

Table 1. Emulsions composition.

AG: Arabic gum; GG: guar gum; EO: essential oil

Microparticles characterization

Lemongrass essential oil microencapsulated analysis

Gas Chromatography With Flame Ionization Detection (GC-FID) was used to analyze the chemical composition of the microencapsulated lemongrass essential oil. 0.2 g of microparticles were added to 5 mL of absolute ethanol and maintained under stirring on a magnetic stirrer for 24 h. The dispersions were filtered in a syringe filter (PVDF membrane, 0.22 µm pore size) and analyzed by GC-FID, using a Hewlett Packard[®] (HP) Series 6850 Gas Chromatograph with a DBS-5 Agilent[®] column (60 m x 0.25 mm). 1µl of the solution was injected, subjected to a chromatographic ramp with a column temperature of 80 °C for 10 min, then raised to 220 °C at 5 °C.min⁻¹. The injector and interface temperature was 250 °C, the split ratio was 1:166, and the carrier gas was nitrogen at 17,52 PSI, with a flow rate of 0.77 mL.min⁻¹ and split the flow of 150 mL.min⁻¹. The components were quantified using an internal pattern and the equipment database.

Microparticles morphology

The morphology and size of the microparticles were evaluated using a field emission gun-scanning electron microscope (FEG-SEM; TESCAN MIRA3). The microparticles were attached to a double-sided adhesive tape, fixed in stubs, and then coated with gold-palladium (Au-Pd) by deposition (sputtering). The FEG-SEM was operated at 10kV with magnifications of 2000-5000x.

Fourier transform infrared spectroscopy (FTIR)

The pure lemongrass essential oil, encapsulating agents, maltodextrin, and the microparticles spectra were acquired by FTIR (IR Prestige 21 - Shimadzu[®]), using a tablet with potassium bromide (KBr), scanned in the spectra range of 4000-400 cm⁻¹, under the analysis conditions of 32 scans.min⁻¹ and 4 cm⁻¹ resolution.

X-ray diffraction (XRD)

The encapsulating agents, maltodextrin, and the microparticles were evaluated by XRD (Ultima IV - Rigaku[®]), using a continuous scan of 2°.min⁻¹, at 20 scanning angles from 5° to 35°, Cu-K α radiation (λ = 1.5418 Å), 30 mA and 40 kV voltage.

Thermogravimetry (TG)

The thermogravimetric curves of pure lemongrass essential oil, encapsulating agents, maltodextrin, and the microparticles were performed using an SDT 2960 instrument (Perkin Elmer[®]), previously calibrated with indium as standard (In; P.F.= 156.6 °C; Δ Hfusion = 28.54 J.g⁻¹). Thermograms were obtained under nitrogen flow (50 mL.min⁻¹) at a constant heating rate of 10 °C.min⁻¹ and temperature range of 20 °C to 550 °C.

RESULTS AND DISCUSSION

Seasonal evaluation of lemongrass essential oil

The seasonal chemical composition of the Lemongrass essential oil (Table 2 and Figure 1) was evaluated. The most applied method for extracting essential oils is hydrodistillation due to the hydrophobic nature of the oil and its lower density than water, allowing the separation of the aqueous phase by decantation [17].

Moreira da Silva, T. L.; et al.

Table 2. Chemica	I composition of	lemongrass es	ssential oil ex	xtracted from le	eaves in the	year's four seasons.
------------------	------------------	---------------	-----------------	------------------	--------------	----------------------

Chemical compounds	AI	Al _{lit}	Spring (%, v/v)	Summer (%, v/v)	Autumn (%, v/v)	Winter (%, v/v)
6-methyl-5-hepten-2-one	983	981	0.56	0.50	1.85	1.01
Myrcene	989	988	3.16	11.81	14.51	14.54
(Z)-β-ocimeno	1035	1032	-	0.24	0.34	0.35
(<i>E</i>)-β-ocimeno	1046	1044	-	-	0.20	0.17
Linalool	1099	1095	1.29	1.06	0.78	0.65
exo-isocitral	1144	1140	0.16	0.25	0.26	0.26
(Z)-isocitral	1161	1160	0.60	0.80	1.02	1.03
(E)-isocitral	1179	1170	1.24	1.65	2.10	2.16
Neral	1238	1235	36.18	33.78	32.02	32.99
Geraniol	1251	1249	3.08	0.69	2.90	1.27
Geranial	1268	1264	48.71	45.61	41.46	43.26
2-undecanone	1292	1293	1.67	-	0.77	0.56
2-tridecanone	1494	1495	-	-	0.20	-
Citral (Neral + Geranial)			84.89	79.39	73.48	76.25
Total identified			96.65	96.39	98.41	98.25

 $AI = calculated arithmetic index; AI_I = arithmetic literature index$

The Brazilian Pharmacopoeia [6] states that the essential oil yield must be 0.5-2.0%. Spring and summer obtained better extraction results (1.96% and 1.95%, respectively), considering the best seasons to obtain higher amounts of the main compounds of the lemongrass essential oil. The amount of essential oil extracted in the autumn and winter was minor (1.50% and 1.48%, respectively); however, all samples were in accordance with the Brazilian Pharmacopoeia specification.

The GC-MS analysis shows that the lemongrass essential oil presents monoterpenes as major constituents, especially the citral, a mixture of two isomeric substances: geranial and neral; the myrcene stands out as another important compound of the mixture, as can be confirmed in Figure 1. The minimum concentration of citral in the lemongrass essential oil is also specificated by the Brazilian Pharmacopoeia (60%) [6]. The citral concentration (sum of the areas of the geranial and neral peaks) in each season was: 84.89% in spring, 79.39% in summer, 73.48% in autumn, and 76.25% in winter, which is in line with the recommendations of the Brazilian Pharmacopoeia.



Figure 1. Chromatograms of lemongrass essential oil extracted in different seasons. A - Winter; B - Autumn; C - Spring; D - Summer. 1 – Myrcene; 2 – Neral; 3 – Geranial.

It was observed that the seasons with higher average temperatures (spring and summer) increase vield and citral production in the lemongrass essential oil, and the myrcene concentration is consequently the opposite, exhibiting low concentration in these seasons. The results agree with studies that showed the best yields of C. citratus essential oil in months of higher temperature; and similar chemical composition, with a predominance of citral as the primary compound, with the lowest percentage in winter, and myrcene with the lowest percentage in spring [18,19]. In conclusion, spring and summer are the best seasons for extracting the lemongrass essential oil due to their higher yield and concentration of citral. Several studies have preferred higher concentrations of citral because it has been proven an essential phytoactive. Citral is used in manufacturing scents, citrus chemicals, cosmetics, food, and pharmaceutical products. Devi and coauthors [20] evidenced the effect of citral extraction of leaves and roots from C. citratus as a vascular smooth muscle relaxant using rat aorta. Bayala and co-authors [21] demonstrated lemongrass essential oil's antioxidant and cytotoxic effects using various prostate cancer and glioblastoma cell lines. The essential oil activity was statistically not different from the isolated citral, concluding that citral is the main responsible for the antiproliferative activity of lemongrass essential oil. Moreover, Long and co-authors [22] showed intense in vivo antibiotic and anti-inflammatory activity, indicating that citral has a strong potential to treat various human and animal diseases.

Microencapsulation of lemongrass essential oil

Due to the small volume obtained from the season extractions, a commercial lemongrass essential oil was used for this study stage. Arabic and guar gums were selected as encapsulating agents since they have emulsifying properties and excellent retention of volatiles. Also, they are natural and food-grade products [23,24]. The gums were combined in different proportions, and it was observed that a higher concentration of guar gum increased the dispersion viscosity, which is inadequate for spray drier use. For this reason, samples AG/GG1:3-EO and GG-EO were not dried. Four samples (AG/GG1:1-EO, AG/GG3:1-EO, AG-EO, and control) generated a powder product.

Lemongrass essential oil microencapsulated analysis

The chemical composition of the microencapsulated analyzed by GC-FID is presented in Table 3. The major components of the lemongrass essential oil were quantified, and the citral concentration in microparticle samples is higher than described in the Brazilian Pharmacopoeia (60%), indicating that the essential oil of lemongrass is present in all samples. The citral concentration of the commercial lemongrass essential oil used in this work is 33.8% of neral and 43.6% of geranial, totaling 77.4% of citral. In this way, the sample composed of arabic and guar gums at 3:1 shows the best encapsulation of the essential oil. The other samples showed decreased citral concentration, indicating that the proportions of wall materials used could not efficiently retain the essential oil, leading to degradation or evaporation during drying.

Compound	AG/GG1:1-EO		AG/GG	3:1-EO	AG-EO	
	r _t	(%)	r _t	(%)	r _t	(%)
Neral	33.75	27.72	33.76	26.98	33.76	27.79
Geranial	34.72	33.59	34.73	45.66	34.72	34.73
Citral (neral + geranial)		61.32		72.65		62.53

Table 3. Retention time (rt) and concentration (%) of the main compounds of lemongrass essential oil after the microencapsulation process obtained by GC-FID.

Microparticles morphology

Figure 2 shows the microparticle images, showing diameters varied from 5 to 20 µm. The powder particles are spherical in shape and smooth, with no significant surface depression, proving the formation of microcapsules. Also, they do not show visible fractures, cracks, or fissures. The presence of cracks on the microcapsule structure is undesirable, as it increases the loss of volatile compounds in a microencapsulated essential oil [25,26,27]. The most pronounced structural difference between the samples was surface topology, specifically the presence or absence of surface collapse. All samples presented agglomeration between the particles, and AG/GG 1:1 showed more collapsed particles.

Different reasons for these particle topologies are discussed, which can occur during drying or storage. The collapse could be attributed to non-encapsulated essential oil deposited on the surface of the microcapsules [13]. The agglomeration also can be due to the drying temperature and feed flow rate that

needs to decrease the superficial tension and viscosity. These parameters can contribute to the formation of droplets and allow the evaporation of liquid before the particles come into contact with the walls of the drying chamber [23]. On the other hand, Bae and Lee [28] relate the agglomeration to the hygroscopicity of wall materials, then after the spray-drying process, the stability of this material is prejudiced. Surface depression or structural collapse and agglomeration of powder particles have been proposed to be related to glass transition and crystallization of amorphous carbohydrates of powder particles. This crystallization is generally thought to occur during powders storage rather than spray drying since spray drying is very rapid, thus forming amorphous particles.



Figure 2. FEG-SEM micrographs of microparticles. (A) AG/GG1:1-EO, (B) AG/GG3:1-EO, (C) AG-EO, (D) AG/GG1:1 (control). First line: 20 µm; 2,000 x; second line: 10 µm; 5,000 x.

Fourier transform infrared spectroscopy (FTIR)

Figure 3 shows FTIR spectra of arabic and guar gums, maltodextrin, lemongrass essential oil, and microparticles. The characteristic peaks of lemongrass essential oil are as follows: 3160 cm⁻¹ (O-H stretch of phenol), 2926 cm⁻¹ (C-H stretch of alkane and chlorophyll group), 1742 cm⁻¹ (C=O stretch of conjugated aldehyde), intense band observed at 1675 cm⁻¹ due to vibrations of C=C (cis and trans), confirming the presence of conjugated double bonds (C=C-CHO) in citral which is common in acyclic monoterpenes, 1378 cm⁻¹ (O-H bend of alcohol), 1040 cm⁻¹ (C-H deformation or C-O/C-C stretching of carbohydrates) [29]. The peak absorption of the O-H group indicated that phenolic compounds appeared at a frequency of 2926 cm⁻¹ [30].



Figure 3. FTIR spectra of lemongrass essential oil, encapsulating agents, and microparticles. (A) over the range 4000-400 cm⁻¹, (B) over the range 3800-2400 cm⁻¹, and (C) over the range 2200-1200 cm⁻¹.

Arabic gum, guar gum, and maltodextrin are polysaccharides, so they have a similar chemical structure based on the glycosidic structure that composes them. Thus, it is observed that all of them presented an absorption band at approximately 3392 cm⁻¹, corresponding to the hydroxyl group -OH; a small band at 2900 cm⁻¹ related to saturated aliphatic C-H groups; stretching at 1640 cm⁻¹ referring to carbonyl C=O; and a stretch between 1000 and 1200 cm⁻¹ where the C-O bonds. Maltodextrin presents a stretch between 1000 and 800 cm⁻¹, corresponding to the pyranose ring of dextroses; guar gum stretching between 700 and 900 cm⁻¹, corresponding to the C-O-C bond between mannose and galactose [13,26,31].

The spectrum of microcapsules was very similar to that of arabic and guar gums, with a slight modification in the band at 2926 cm⁻¹ referent to the O-H group of phenolic compounds of essential oil that reduced its intensity in the microparticle spectra (Figure 3b). The absorption band at 1675 cm⁻¹ was used as a reference to assess the presence of lemongrass essential oil in microparticles. All formulations were confirmed to contain lemongrass essential oil, as shown by the presence of the absorption band in the enlarged spectral region (Figure 3c); however, there was a decrease in intensity, confirming the successful encapsulation and formation of microcapsules. The carbonyl vibrations of the gums reduced the band intensity at 1640 cm⁻¹ in the microparticle samples containing the lemongrass essential oil (Figure 3c). Other spectral changes may be due to the overlapping of lemongrass oil and polysaccharides in a similar spectra range. The essential oils can modify intermolecular interactions in the polymer [29], and it infers the presence of lemongrass oil on the microcapsules.

X-ray diffraction (XRD)

The XRD characterization has determined the materials' nature as crystalline or amorphous. Powders with crystalline states present well-ordered regions with sharp and defined peaks, while amorphous products produce a broad peak due to their disordered state [13]. The XRD diffractogram of the powders is shown in Figure 4. For individual encapsulation agents, arabic gum exhibited one broad peak at 2θ of 18°, and guar gum shows any crystalline region defined, demonstrating an amorphous structure. On the other hand,

maltodextrin diffractogram presents several well-defined peaks, more intense at 20 of 12°, 13°, 19°, and 25°, indicating its high crystallinity.



Figure 4. XRD diffractogram of (A) microparticles and encapsulating agents; (B) maltodextrin.

All microparticles showed an amorphous structure, but some crystallinity peaks at 20 were identical to the diffractogram of maltodextrin. It occurs because the maltodextrin was added before the drying process as a cryoprotectant and not as an encapsulating material; therefore, it is not incorporated into the microparticle, becoming dispersed or adhered to the outer part of the wall of the microparticles, generating the corresponding peaks. These finds are consistent with previous studies results, which reported amorphous structures in microparticles obtained by spray-drying [13,32,33].

Thermogravimetry (TG)

TG curves of lemongrass essential oil, encapsulating agents, and microparticles are depicted in Figure 5.



Figure 5. TG curves of lemongrass essential oil, encapsulating agents, and microparticles.

TG curve of lemongrass essential oil shows a first stage of thermal decomposition with T_{onset} at 107 °C and a second event with T_{onset} at 359 °C. Arabic and guar gums and maltodextrin had more than one stage of decomposition. The first mass loss of polysaccharides was probably related to moisture loss, and a second event with T_{onset} at 236, 240, and 203 °C, respectively.

The thermogravimetric analysis allowed for estimating the T_{max} , the maximum mass loss rate temperature. The T_{max} of the first mass loss event of lemongrass essential oil was 210 °C, and that of arabic and guar gums was 61 and 53 °C. Microparticles containing the lemongrass essential oil had a T_{max} of 109, 117, and 127 °C, and the control microparticle had a T_{max} of 50 °C, corresponding to the first mass loss event. Differences in values show that the loss is predominantly associated with water rather than oil.

Regarding the second mass loss event, the TG curves of microparticles show T_{onset} at 184, 191, and 194 °C in samples containing the lemongrass essential oil (AG/GG1:1-EO, AG/GG3:1-EO, and AG-EO, respectively), and 187 °C in microparticle without the essential oil (AG/GG1:1; control). Because these values are higher than those observed for free lemongrass essential oil (107 °C), it can be inferred that microencapsulation improved the thermal stability of all formulations. Complementary TG curves show a mass loss event between 185 and 226 °C that is not typical of free essential oil or encapsulants, suggesting oil volatilization throughout a larger temperature range. This result confirms the effectiveness of microencapsulation and the correct choice of encapsulants [34,35,36,37,38].

Also, it is possible to notice that the second stage of mass loss in all the microcapsules starts near the temperature of maltodextrin, evidencing its efficiency as a cryoprotectant.

CONCLUSION

This study showed that seasonality greatly influences the yield and chemical composition of the lemongrass essential oil, showing better results in the seasons with higher average temperatures, spring and summer. Citral (neral and geranial mixture) and myrcene were the major compounds in the extraction identified by the GC-MS method. Microencapsulation of lemongrass essential oil by a spray-drying technique using arabic and guar gums produced a fine powder, showing agglomeration and collapse particles with no apparent pores or cracks, essential to incorporate volatile compounds. In addition, the characterization techniques indicated the lemongrass essential oil encapsulation in amorphous particles, improving its thermal

stability and maintaining high citral concentration. Also, using maltodextrin as a drying aid improved the stability of the microparticles. Then, more studies relating to the stability and application of the microcapsules are necessary to confirm the microencapsulation efficacy in food products, drugs, or cosmetics.

Funding: This work was supported by Coordenadoria de Aperfeiçoamento de Pessoal do Ensino Superior (CAPES). **Acknowledgments:** We thank Geroma do Brasil Indústria e Comércio Ltda for the GC-FID support, Quinari/Yanih Produtos Cosméticos e Farmacêuticos for the donation of lemongrass essential oil, and c-LABMU/UEPG for physicochemical analysis support.

Conflicts of Interest: The authors declare no conflict of interest.

REFERENCES

- 1. Croteau R, Kutchan TM, Lewis NG. Natural products. In: Biochemistry and molecular biology of plants. 2nd ed. Hoboken (NJ): John Wiley and Sons; 2015. p. 1132–205.
- Gobbo-Neto L, Lopes NP. Medicinal plants: factors of influence on the content of secondary metabolites. Quim Nova [Internet]. 2006 Oct;30(2):374-81. Available from:
- https://www.scielo.br/j/qn/a/gn5mhqcFHSbXXgTKNLJTS9t/?format=pdf&lang=pt.
- 3. Turek C, Stintzing FC. Stability of essential oils: A review. Compr Rev Food Sci Food Saf. 2013 Jan;12(1):40–53. doi:10.1111/1541-4337.12006.
- Aćimović M, Čabarkapa I, Cvetković M, Stanković J, Kiprovski B, Gvozdenac S, et al. *Cymbopogon citratus* (DC.) Staph: Chemical composition, antimicrobial and antioxidant activities, use in medicinal and cosmetic purpose. J Agron Technol Eng Manag [Internet]. 2019 Nov; 2(6):344-60. Available from: https://fiver.ifvcns.rs/handle/123456789/2219.
- 5. Sousa R, Figueirinha A, Batista MT, Pina ME. Formulation effects in the antioxidant activity of extract from the leaves of *Cymbopogon citratus* (DC) Stapf. Molecules. 2021 July;26(15):1-15. doi:10.3390/molecules26154518.
- 6. Brazilian Health Surveillance Agency. Plantas Medicinais [Medicinal Plants]. Ph. Braz. 6th ed. Brasília (Brazil): 2019; 746 p.
- Majewska E, Kozlowska M, Gruczynska-Sekowska E, Kowalska D, Tarnowska K. Lemongrass (*Cymbopogon citratus*) essential oil: Extraction, composition, bioactivity and uses for food preservation a review. Pol J Food Nutr Sci. 2019 Nov;69(4):327-41, doi:10.31883/pjfns/113152.
- 8. Falleh H, Jemaa MB, Saada M, Ksouri R. Essential oils: A promising eco-friendly food preservative. Food Chem. 2020 Nov;330: 1–40, doi:10.1016/j.foodchem.2020.127268.
- 9. Eslahi H, Fahimi N, Sardarian AR. Chemical composition of essential oils. In: Essential oils in food processing: Chemistry, safety and applications. 1. ed. Oxford (UK): John Wiley and Sons, 2018. p. 119-71.
- 10. Aguiar MCS, Silva MFGF, Fernandes JB, Forim MR. Evaluation of the microencapsulation of orange essential oil in biopolymers by using a spray-drying process. Sci Rep. 2020 July;10:1–11. doi:10.1038/s41598-020-68823-4.
- 11. Pereira KC, Ferreira DCM, Alvarenga GF, Pereira MSS, Barcelos MCS, Costa JMG. Microencapsulation and release controlled by the diffusion of food ingredients produced by spray drying: a review. Braz J Food Technol. 2018 June;21:1–9. doi:10.1590/1981-6723.08317.
- 12. Oxley J. Overview of microencapsulation process technologies. In: Microencapsulation in the food industry: A practical implementation guide. [place unknown]: Elsevier; 2014. p. 35-46.
- Kang Y, Lee Y, Kim YJ, Chang YH. Characterization and storage stability of chlorophylls microencapsulated in different combination of gum arabic and maltodextrin. Food Chem. 2019 Jan;272:337-46. doi:10.1016/j.foodchem.2018.08.063.
- Castañeda-Óvando A, González-Aguilara LA, Granados-Delgadilloa MA, Chávez-Gómez UJ. Guar Gum: an allied in food industry. Pädi Boletín Científico De Ciencias Básicas E Ingenierías Del ICBI. 2020 May;7(14):107-11. doi:10.29057/icbi.v7i14.4988.
- Xiao A, Kang Y, Hou W, Niu Y, Kou X. Microcapsules based on octenyl succinic anhydride (OSA)-modified starch and maltodextrins changing the composition and release property of rose essential oil. Int J Biol Macromol. 2019 Sept;137:132–38. doi:10.1016/j.ijbiomac.2019.06.178.
- 16. Adams RP. Identification of essential oil components by gas ghromatography/quadrupole mass spectroscopy. 4th ed. Carol Stream (IL): Allured Pub Corp; 2007. 804 p.
- 17. Asbahani AE, Miladi K, Badri W, Sala M, Addi EHA, Casabianca H, et al. Essential oils: From extraction to encapsulation. Int J Pharm. 2015 April;483(1-2):220-43. doi:10.1016/j.ijpharm.2014.12.069.
- 18. Santos A, Paduan RH, Gazin ZC, Jacomassi E, D'Oliveira PS, Cortez DAG, et al. Assessment of the yield and antimicrobial activity of *Cymbopogon citratus* (DC.) Stapf essential oil in relation to the seasonality and cultivation in consortium." Rev. bras. farmacogn. 2009 June;19(2a):436-41. doi:10.1590/S0102-695X2009000300017.
- Madi YF, Choucry MA, Meselhy MR, El-Kashoury EA. Essential oil of *Cymbopogon citratus* cultivated in egypt: seasonal variation in chemical composition and anticholinesterase activity. Nat. Prod. Res. 2021 Nov;35(21):4063-67. doi:10.1080/14786419.2020.1713125.
- 20. Devi RC, Sim SM, Ismail R. Effect of *Cymbopogon citratus* and citral on vascular smooth muscle of the isolated thoracic rat aorta. Evid Based Complementary Altern Med. 2012 May;2012:1–8. doi:doi.org/10.1155/2012/539475.

- 21. Bayala B, Bassole IHN, Maqdasy S, Baron S, Simpore J, Lobaccaro JA. *Cymbopogon citratus* and *Cymbopogon giganteus* essential oils have cytotoxic effects on tumor cell cultures. Identification of citral as a new putative antiproliferative molecule. Biochimie. 2018 Oct;153:162-70. doi:10.1016/j.biochi.2018.02.013.
- 22. Long N, Tang H, Sun F, Lin L, Dai M. Effect and mechanism of citral against Methicillin-Resistant *Staphylococcus aureus* in vivo. J Sci Food Agric. 2019 Mar;99(9):4423-29. doi:10.1002/jsfa.9677.
- 23. Veiga RS, Silva-Buzanello RA, Corso MP, Canan C. Essential oils microencapsulated obtained by spray drying: a review. J Essent Oil Res. 2019 May;31(6):457–73. doi:10.1080/10412905.2019.1612788.
- 24. Bakry AM, Abbas S, Ali B, Majeed H, Abouelwafa MY, Mousa A, et al. Microencapsulation of oils: a comprehensive review of benefits, techniques, and applications. Compr Rev Food Sci Food Saf. 2016 Jan;15(1):143-82. doi:10.1111/1541-4337.12179.
- Fernandes RVB, Borges SV, Botrel DA, Silva EK, Costa JMG, Queiroz F. Microencapsulation of rosemary essential oil: characterization of particles. Dry. Technol. 2013 Aug;31(11):1245-54. doi:10.1080/07373937.2013.785432.
- 26. Mehran M, Masoum S, Memarzadeh M. Microencapsulation of Mentha spicata essential oil by spray drying: optimization, characterization, release kinetics of essential oil from microcapsules in food models. Ind Crops Prod. 2020 Oct;154:1-8. doi:10.1016/j.indcrop.2020.112694.
- 27. Burhan AM, Abdel-Hamid SM, Soliman ME, Sammour OA. Optimisation of the microencapsulation of lavender oil by spray drying. Journal of Microencapsulation. 2019 June;36(3):250-66. doi:10.1080/02652048.2019.1620355.
- 28. Bae EK, Lee SJ. Microencapsulation of avocado oil by spray drying using whey protein and maltodextrin. J Microencapsul. 2008 Oct;25(8):549-60. doi:10.1080/02652040802075682.
- 29. Gogoi M, Kadam V, Jose S, Shakyawar DB, Kalita B. Multifunctional finishing of woolens with lemongrass oil. J Nat Fibers. 2020 May;19(4):1353-65. doi:10.1080/15440478.2020.1764458.
- Sathiyamoorthi R, Sankaranarayanan G. Effect of antioxidant additives on the performance and emission characteristics of a DICI engine using neat lemongrass oil-diesel blend. Fuel. 2016 June;174:89-96. doi:10.1016/j.fuel.2016.01.076.
- Başyiğit B, Sağlam H, Kandemir Ş, Karaaslan A, Karaaslan M. Microencapsulation of sour cherry oil by spray drying: Evaluation of physical morphology, thermal properties, storage stability, and antimicrobial activity. Powder Technol. 2020 Mar;364:654-63. doi:10.1016/j.powtec.2020.02.035.
- Ban Z, Zhang J, Li L, Luo Z, Wang Y, Yuan Q, et al. Ginger essential oil-based microencapsulation as an efficient delivery system for the improvement of Jujube (*Ziziphus jujuba* Mill.) fruit quality. Food Chem. 2020 Feb;306:1-34. doi:10.1016/j.foodchem.2019.125628.
- Nambiar RB, Sellamuthu PS, Perumal AP. Microencapsulation of tender coconut water by spray drying: Effect of Moringa oleifera gum, maltodextrin concentrations, and inlet temperature on powder qualities. Food Bioproc Tech. 2017 June;10(9):1668-84. doi:10.1007/s11947-017-1934-z.
- Melo AM, Barbi RCT, Souza WFC, Luna LC, Souza HJB, Lucena GL, et al. Microencapsulated lemongrass (*Cymbopogon Flexuosus*) essential oil: a new source of natural additive applied to coalho cheese. J Food Process Preserv. 2020 Oct;44(10):1-32. doi:10.1111/jfpp.14783.
- 35. Mazuco RA, Cardoso PMM, Bindaco ES, Scherer R, Castilho RO, Faraco AAG, et al. Maltodextrin and gum arabic-based microencapsulation methods for anthocyanin preservation in Juçara palm (*Euterpe edulis martius*) fruit pulp. Plant Foods Hum Nutr. 2018 June;73(3):209-15. doi:10.1007/s11130-018-0676-z.
- 36. Barbieri N, Sanchez-Contreras A, Canto A, Cauich-Rodriguez JV, Vargas-Coronado R, Calvo-Irabien LM. Effect of cyclodextrins and mexican oregano (*Lippia graveolens kunth*) chemotypes on the microencapsulation of essential oil. Ind Crops Prod. 2018 Oct;121:114-23. doi:10.1016/j.indcrop.2018.04.081.
- 37. Felix PHC, Figueiredo JA, Marque GR, Botrel DA, Borges SV. Termogravimetria (TGA) de micropartículas de óleo essencial de canela. [Thermogravimetry (TGA) of cinnamon essential oil microparticles]. Proceedings of the 24th UFLA Postgraduate Congress; 2015; Lavras (MG, Brazil). Federal University of Lavras, 2015.
- 38. Martins WS, Araújo JSF, Feitosa BF, Oliveira JR, Kotzebue LRV, Agostini DLS, et al. Lemongrass (*Cymbopogon citratus* D.C. Stapf) essential oil microparticles: development, characterization, and antioxidant potential. Food Chem. 2021 Sep;355:1-7. doi:10.1016/j.foodchem.2021.129644.



© 2023 by the authors. Submitted for possible open access publication under the terms and conditions of the Creative Commons Attribution (CC BY NC) license (https://creativecommons.org/licenses/by-nc/4.0/).