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Optimization of operating parameters in extraction of essential oils from *Lavandula angustifolia* flowers by microwave assisted steam distillation

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ABSTRACT

The aim of the study to optimize the operating parameters on the yield, composition, time and kinetics in extraction of essential oils from different *Lavandula angustifolia* flowers by microwave assisted steam distillation (MSD) in comparison to conventional steam distillation (SD). A preliminary study allowed for the determination of the most influential operating factors, namely water vapor flow rate and microwave heating power. The yield obtained by MSD was comparable to that obtained by SD, while the extraction time was considerably reduced with 6 min for MSD compared to 30 min for SD. Optimization of the operating parameters was assessed using response surface methodology (RSM). The optimal treatment conditions obtained were: 8 g·min^{-1} for the steam flow rate, 500 W for the microwave power and 6 min for the extraction time. The main compounds identified in the different lavender essential oils extracted by MSD chosen as the variables in a multivariate study. A composite centered design based on a response surface methodology was investigated for each major constituent found in the oils. The results showed that the chemical composition of the lavender essential oil was strongly influenced by the MSD operating conditions. The choice of particular operating parameters will allow for extraction of a specific compound.

Keywords: Microwave steam distillation; *Lavandula angustifolia* Mill.; Response surface methodology

1. Introduction

In the industrial field, it is often a question of first identifying the most influential factor(s) in a manufacturing process because for a system under study, there can be an almost unlimited number of variables involved in the design. To choose these factors, it is necessary to have a minimum of prior knowledge of the phenomenon under study. This knowledge can be acquired through a literature review and/or extended through preliminary experiments to set certain parameters [1]. Experimental designs allow for a better understanding of the phenomena involved in new processes developed and make it possible to grasp a response variable quickly. The experimental design theory was born out of a multitude of practical problems in all science fields where the need to organize an experiment to improve the results – without increasing the cost – was indispensable. Today, many authors deal with the use of experimental designs in various fields with the aim of studying, optimizing, comparing, or estimating various criteria [2,3].

The central composite design (CCD) method was first proposed by Box and Wilson [4] and more recently by Yoon [5] and Goupy [6]. The method consists of full or fractional factorial designs and allow an efficient estimation of a second degree model by assigning more than 2 levels to the

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factors. In a centered composite design, tests were added to the center and outside of the domain.

The aim of the study was to optimize the operating parameters on the yield, composition, time, and kinetics in extraction of essential oils from *Lavandula angustifolia* flowers by microwave assisted steam distillation (MSD) in comparison to conventional steam distillation (SD). A composite centered design based on a response surface methodology was investigated for each major constituent found in the oils.

2. Materials and methods

2.1. Plant material

Lavender flowers (*Lavandula angustifolia* Mill., Lamiaceae) were collected in July 2019 and left to dry in the shade.

2.2. Essential oil extraction

Lavender flower essential oil was recovered by microwave steam distillation (MSD) and conventional steam distillation (SD). Essential oil yield (Y_{EO}) was estimated according to dry vegetable matter and expressed in g of essential oil (EO) per 20 g of dry matter.

2.2.1. Microwave steam distillation

The process was based on the principle of conventional steam entrainment (SD) in which microwave irradiation was applied to the extraction reactor inside the microwave oven. The refrigeration system and part of the essential oil recovery system was placed outside the microwave oven. The resulting oil was steamed into the refrigeration system where it was condensed and then collected in a Florentine vessel. It was separated from the water by decantation, dried on sodium sulphate and stored at 4°C [7,8].

2.2.2. Steam distillation apparatus and procedure

For a rigorous comparison, the same glassware and same operating conditions were used for conventional SD [9]. The vapor produced by the steam generator crossed the plant charged with essential oil and then passed through a condenser to a receiving Florentine flask. The essential oil was collected, dried with anhydrous sodium sulfate, and stored at 4°C until use. Extractions were performed at least three times, and the mean values were reported.

2.3. Analysis of essential oils

Essential oils obtained by both MSD and SD were analyzed by gas chromatography coupled to mass spectrometry (GC/MS) using a Hewlett-Packard computerized system comprising a 6890-gas chromatograph coupled to a 5973A mass spectrometer. A fused silica-capillary column HP5MS (30 m × 0.25 mm, 0.25 μm film thickness) was employed. GC/MS spectra were obtained using the following conditions: carrier gas He; flow rate 0.3 mL·min^{-1}; splitless mode; injection volume 1 μL; injection temperature 250°C; oven temperature program was 60°C for 8 min, increasing at 2°C/min to 250°C and held at 250°C for 15 min. The ionization mode used was electron impact at 70 eV. The relative percentage of the components was calculated from GC by flame ionization detection (GC–FID). Most constituents were identified by comparison of their GC Kovats retention indices (RI), determined with reference to a homologous series of C9-C17 *n*-alkanes. Some structures were further confirmed by authentic standards available in the author's laboratory analyzed under the conditions described above. Identification was confirmed when possible; by comparison of their mass spectral fragmentation patterns with those stored in the MS database and with mass spectra literature data [10,11].

2.4. Optimization

2.4.1. Preliminary study

A series of preliminary experiments were conducted and to determine factors that particularly influenced the extraction kinetics of the essential oil. The mass of plant material was set at 20 g for all trials. The yield of essential oil was calculated on a dry matter basis. The plant material was air-dried and protected from the sun.

2.4.2. Experimental design

A Box–Wilson procedure, also known as central composite design (CCD), was used to achieve maximal information of the process with a minimal number of possible experiments. The multivariate study allowed for the identification of interaction between variables and provided a complete assessment of the experimental domain. The type of CCD used was a central composite face centered (CCF) experimental design to determine the optimal conditions of the MSD process. The application of a CCF design was a convenient way to optimize a five-level process (–α,–1, 0, +1, +α) for each factor. In this design, the star points were at the center of each face of the factorial space, thus ±α = ±1 [6,12]. A total of 18 different combinations, including 2^3 full factorial designs (±1) with four axial points (±α) and four replicates of center point (coded 0), were investigated to fit the full quadratic equation model given by Eq. (1):

$$Y_{EO} = \beta_0 + \sum_{i=1}^{k}\beta_i X_i + \sum_{i=1}^{k}\beta_{ii} X_i^2 + \sum_{i=1}^{k-1}\sum_{j=i+1}^{k}\beta_{ij} X_i X_j \quad (1)$$

where Y_{EO} represented the response variable (essential oil yield in this study), β_0, β_i, β_{ii}, and β_{ij} were the regression coefficients of variables for intercept, simple, quadratic and interaction terms, respectively. X_i and X_j were the independent coded variables influencing the response variable Y_{EO}, and k represented the quantity of variables. The range of parameters was chosen based on a parametric analysis (Table 1). The results obtained from the CCD were analyzed statistically with software Statgraphics plus (Version 5.1, Statistical Graphics Corporation, Rockville, USA, 2000).

3. Results and discussion

3.1. Extraction yield and time

Variation in yields of *Lavandula angustifolia* Mill., Lamiaceae as a function of time (Fig. 1) showed differences

Table 1
Areas of operating parameters variation

Factors	Levels				
	–α	–1	0	+1	+α
Steam flow rate (g·min^{-1})	4	4	8	12	12
Power (W)	300	300	500	700	700

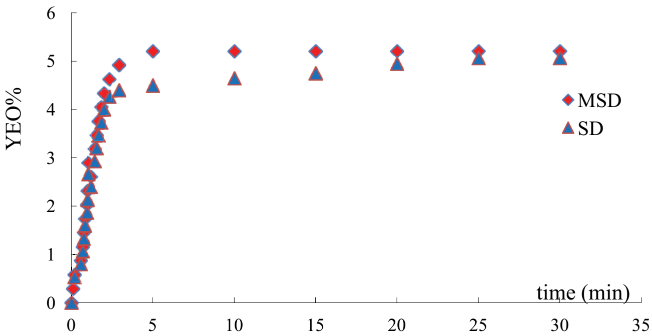

Fig. 1. Profile of essential oil yield obtained by MSD and SD as a function of extraction time.

in the kinetics of the two processes. The microwave-assisted steam extraction was much faster; 5 min were enough to extract 5.21% of essential oil, whereas it took 25 min to extract 5.07% by conventional steam extraction.

3.2. Preliminary study

Initial studies indicated that the steam flow rate and the microwave heating power were key factors in the extraction process. Based on this the steam flow rate (2, 4, 6, 8 and 12 g·min^{-1}) and microwave power (200, 300, 400, 500 and 700 W) were optimized (Table 2). A high flow rate may have suggested the creation of preferential pathways, consequently decreasing the contact time between the steam and the lavender flowers and thus reducing the yield of essential oil. A low flow rate may not have been sufficient to extract all the essential oil. The optimal extraction yield (5.07%) was obtained for a steam flow rate of 8 g·min^{-1}. Furthermore, different microwave irradiation powers were examined for MSD extraction of lavender flower essential oil with an optimal steam flow rate (8 g·min^{-1}). The power should not have been too high, in order not to lose volatile compounds. The optimal extraction yield of 5.21% was obtained at 500 W.

It should be remembered that this method only made it possible to evaluate the individual effect of each of the parameters on the yield without taking into account their interactions, hence the need for a study by experimental design.

3.3. Central composite design results

After having selected the main operating parameters and explained their relevant range of variation, thanks to the preliminary tests, we opted for the centered composite design method to optimize the MSD extraction process. The trials were randomized to minimize the effects of unexpected variability in responses due to unrelated

Table 2
Results of preliminary study

Q (g·min^{-1})	Yield (%)	P (W)	Yield (%)
2	5.06	200	4.63
4	4.79	300	4.72
6	4.55	400	4.79
8	5.07	500	5.21
12	4.17	700	4.94

factors. The experiments were performed using the operating conditions described in (Table 3).

According to Tables 2 and 3, the optimal performance was obtained for the centre point tests which corresponded to the values of the coordinate parameters: coded variables (0,0) and real variables (Q = 8 g/min and P = 500 W). Furthermore, the results of the central composite design were analyzed by STATGRAPHICS® software. The purpose of this analysis was to determine the main effects of each parameter, their quadratic effects, the interactions between them and the development of the empirical model describing the behavior of the system studied.

3.4. Analysis of variance

The effects of the parameters were checked graphically using the Pareto-chart, which showed the standardized effects in Fig. 2. Those effects were significant if they exceeded the value symbolized by the vertical line with a risk of error of 5%. It should be remembered that the standardized effect was expressed as the observed effect divided by the estimated standard deviation of the effect. The results of the analysis of variance (ANOVA) showed that the most significant effect was steam flow rate. Among the quadratic effects, power was the most significant followed by that of steam ($p < 0.05$).

Statistical analysis of the results by the CCD's STATGRAPHICS® software allowed for determination of an empirical relationship between the response studied (Y_{EO}) and the key variables involved in the model, which was described by the following polynomial equation of the fitted model [Eq. (2)].

$$Y_{EO} = 5.16833 - 0.388333A - 0.005B - 0.28AA + 0.0025 - 0.3BB \quad (2)$$

where Y_{EO} (%): essential oil yield, Q: steam flow (g·min^{-1}), P: microwave power (W).

The coefficients of this model expressed the effect of the variables on the essential oil yield. A positive value supported the operation, whereas a negative value did not. The regression coefficient indicated a goodness of fit of the estimates of the regression equation R^2. It was equal to 94.527%, which meant that the model described the system well. The response surfaces allowed for represention of the total effect of operative parameters. They were used to search for experimental regions for which the response was optimal. Fig. 3 representing the variation of the essential oil yield as a function of the flow-power couple combined

Table 3
Results of central composite design for (*Lavandula angustifolia* Mill., Lamiaceae) essential oil extracted by MSD

Runs	Real variable		Coded variable		Yield (%)
	Q (g·min^{-1})	P (W)	A	B	
1	4	300	–1	–1	5.12
2	12	300	1	–1	4.22
3	4	700	–1	+1	4.99
4	12	700	1	1	4.10
5	4	500	–1	0	5.12
6	12	500	+1	0	4.58
7	8	300	0	–1	4.71
8	8	700	0	+1	4.94
9	8	500	0	0	5.21
10	8	500	0	0	5.19
11	8	500	0	0	5.17
12	8	500	0	0	5.18

Note: The values in bold and underlined mean correspond to the optimal conditions obtained by RSM study.
[a]Exp.: Experimental value of essential oil yield.
[b]Pred.: Predicted value of essential oil yield.

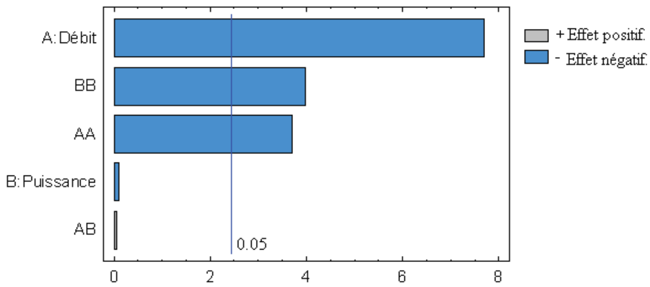

Fig. 2. Pareto chart.

effects shows that the optimal yield was located in the zone of space corresponding to the values of the experimental domain centre with coordinates (0, 0) in reduced variables and (8 g·min^{-1}, 500 W) in real variables

3.5. Results of the experimental design study of major compounds relative contents of lavender essential oil extracted by MSD

The peak areas of the main constituents, identified in the different lavender essential oils extracted by MSD, were considered as response variables in a multivariate study. Thus, a composite centered design based on a response surface methodology was investigated for each major constituent in the different oils. The results of the GPC analyses of the MSD extracted lavender oil samples for the majority constituents corresponding to the 12 trials programmed by the centred composite design are given in Table 4 and the analysis of variance values in Table 5. Examination of Table 4 shows that the chemical composition of the lavender essential oil was strongly influenced by the operating conditions of the microwave-assisted steam distillation. Indeed, the relative contents of the majority compounds

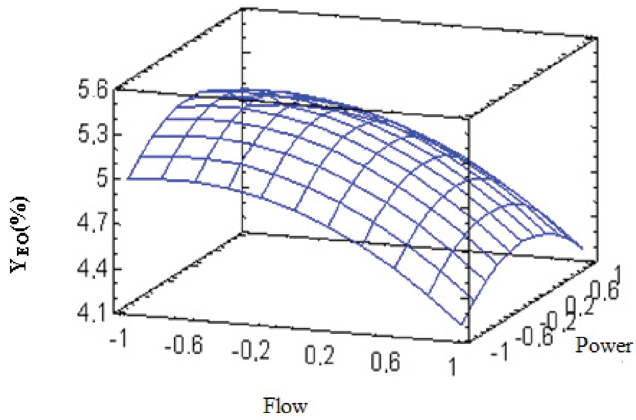

Fig. 3. Response surface corresponding to the flow-power pair.

varied from one trial to another. Based on these results it was possible to choose the experimental extraction conditions that will allow for making a specific extraction of one compound compared to another.

Analysis of variance (Table 6) was performed on the results. This made it possible to test and evaluate the statistical significance of each of the effects of the parameters (i.e., linear, quadratic and interaction). The linear effect of flow rate significantly influenced the content of 1,8-cineole, camphor and linalyl acetate. The combined effect of flow rate and power had a significant effect on borneol content. None of the factors seemed to have an influence on linalool and terpinen-4-ol. The analysis of the results by the STATGRAPHICS® software allowed for the proposing of an empirical model in real and reduced coordinates for each compound.

- Coded variables
  1,8-cineole % = 4.21138 – 0.823667A + 0.244333B + 0.289375AA + 0.38825AB – 0.648625BB
  Linalool % = 30.0274 + 0.232167A – 0.314333B + 0.526875AA – 0.658AB + 1.18738BB
  Camphor % = 7.91812 – 0.335667A + 0.117667B + 0.001125AA – 0.281AB – 0.411875BB
  Borneol % = 5.12108 – 0.0566667A – 0.108833B – 0.09325AA – 0.36675AB – 0.05775BB
  Linalyl acetate % = 25.5558 + 0.737333A + 0.377B – 0.559875BB + 0.53875AB + 0.284125BB
  Terpinen-4-ol % = 5.36771 + 0.190167A – 0.00966667B + 0.416875AA – 0.00625AB + 0.429375BB
- Real variables
  1,8-cineole % = 4.29272 – 0.737948 Flow + 0.0135548 Power + 0.0180859 Flow-Flow + 0.000485312 Flow-Power – 0.0000162156 Power-Power
  Linalool % = 36.5875 – 0.0575833 Flow – 0.024676 Power + 0.0329297 Flow-Flow – 0.0008225 Flow-Power + 0.0000296844 Power–Power
  Camphor % = 4.32057 + 0.0905833 Flow + 0.0136952 Power + 0.0000703125 Flow-Flow – 0.00035125 Flow-Power – 0.0000102969 Power-Power
  Borneol % = 2.93881 + 0.308302 Flow + 0.00456708 Power – 0.00582812 Flow-Flow – 0.000458437 Flow-Power – 0.00000144375 Power-Power

Table 4
Variation of main compounds relative contents of lavender essential oils according to operating conditions

Runs	Flow (g·min⁻¹)	Power (W)	1,8-cineole %	Linalool %	Camphor %	Borneol %	Linalyl acetate %	Terpinen-4-ol %
1	–1	–1	5.055	31.24	7.657	4.810	24.455	5.835
2	1	–1	2.855	32.668	7.586	5.369	26.456	6.363
3	–1	+1	4.684	30.981	8.257	5.208	24.552	5.718
4	1	1	4.037	29.777	7.062	4.300	26.708	6.221
5	–1	0	4.937	31.120	8.027	5.120	24.337	6.089
6	+1	0	2.842	32.289	7.279	5.129	26.604	6.199
7	0	–1	2.624	31.733	6.925	5.151	26.358	6.056
8	0	+1	3.279	32.997	7.555	5.169	26.271	6.257
9	0	0	4.502	29.678	8.008	5.056	25.197	4.997
10	0	0	4.570	29.045	8.022	5.049	25.411	5.301
11	0	0	4.626	29.682	8.149	5.120	25.222	5.272
12	0	0	4.370	29.404	8.026	5.066	25.444	5.182

Table 5
Results of analysis of variance of major constituents of lavender essential oils

Response variable	Val-P					
	1,8-cineole	Linalool	Camphor	Borneol	Linalyl acetate	Terpinen-4-ol
A: Flow	0.0196	0.6926	0.0359	0.3490	0.0290	0.2798
B: Power	0.3851	0.5946	0.3817	0.0990	0.1946	0.9538
AA	0.4875	0.5533	0.9954	0.3078	0.1985	0.1332
AB	0.2699	0.3740	0.1153	0.0017	0.1394	0.9756
BB	0.1484	0.2069	0.0699	0.5160	0.4909	0.1240
R^2	71.5821	44.2965	73.88	85.76	71.8885	64.13

Table 6
Experimental and theoretical optimal contents of the majority compounds of the essential oil extracted by MSD

Optimal content	1,8-cineole	Linalool	Camphor	Borneol	Linalyl acetate	Terpinen-4-ol
Experimental	5.055	32.997	8.257	5.369	26.708	6.363
Theoretical	4.819	30.900	8.241	5.389	26.933	6.420

Linalyl acetate % = 25.3687 + 0.40749 Flow – 0.0106056 Power – 0.0349922 Flow-Flow + 0.000673438 Flow-Power + 0.00000710313 Power-Power

Terpinen-4-ol % = 9.33139 – 0.365427 Flow – 0.0107202 Power + 0.0260547 Flow-Flow – 0.0000078125 Flow-Power + 0.0000107344 Power-Power

4. Conclusions

The extraction of essential oil from lavender flowers by (MSD) and (SD) was successfully assessed quantitatively (yield) and qualitatively (chemical composition). The essential oils isolated by these two processes were similar. However, the MSD process was more efficient with a considerable gain in extraction time (6 min vs. 30 min) and consequently in energy consumption. A parametric study was conducted to evaluate the influence of microwave heating power and steam flow rate. This allowed for the evaluation of the individual effects of the operating parameters on the essential oil yield and the definition of the experimental area for the design study.

The trials programmed by the centered composite design allowed for estimation of the individual, quadratic and conjugate effects of the operating parameters on the essential oil yield. A mathematical model describing the variation in yield as a function of these parameters was proposed. It was found that the individual and quadratic effects of the steam flow rate were positive, while those of the heating power and the interaction effects of the two parameters were negative.

The GC analyses of the samples of lavender essential oil extracted by MSD for the majority constituents corresponding to the 12 trials programmed by the central composite plan, showed that the chemical composition of lavender essential oil was influenced by the operating conditions. Indeed, the relative contents of the major compounds varied from one trial to another, which allowed for the out of selective extractions of a given compound. By choosing optimal experimental conditions it was possible to obtain a high percentage of essential oil.

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