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Short communication

Improved microwave steam distillation apparatus for isolation of essential oils Comparison with conventional steam distillation

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ABSTRACT

Steam distillation (SD) is routinely used by analysts for the isolation of essential oils from herbs, flowers and spices prior to gas chromatographic analysis. In this work, a new process design and operation for an improved microwave steam distillation (MSD) of essential oils from aromatic natural products was developed. To demonstrate its feasibility, MSD was compared with the conventional technique, SD, for the analysis of volatile compounds from dry lavender flowers (*Lavandula angustifolia* Mill., Lamiaceae). Essential oils isolated by MSD were quantitatively (yield) and qualitatively (aromatic profile) similar to those obtained by SD, but MSD was better than SD in terms of rapidity (6 min versus 30 min for lavender flowers), thereby allowing substantial savings of costs in terms of time and energy. Lavender flowers treated by MSD and SD were observed by scanning electron microscopy. Micrographs provide evidence of more rapid opening of essential oil glands treated by MSD, in contrast to conventional SD.

1. Introduction

With the increasing energy prices and the drive to reduce CO_2 emissions, universities and industries are challenged to find new technologies in order to reduce energy consumption, to meet legal requirements on emissions, and for cost reduction and increased quality. For example, existing extraction technologies have considerable technological and scientific bottlenecks to overcome: often requiring more than 70% of total process energy used. Driven by these goals, advances in microwave extraction have resulted in a number of techniques such as microwave-assisted solvent extraction (MASE) [1,2], vacuum microwave hydrodistillation (VMHD) [3], microwave hydrodistillation (MWHD) [4], compressed air microwave distillation (CAMD) [5], solvent-free microwave extraction (SFME) [6], microwave hydro-diffusion and gravity (MHG) [7] and microwave-accelerated steam distillation (MASD) [8]. Over the years procedures based on microwave extraction have replaced some of the conventional processes and other thermal extraction techniques that have been used for decades in chemical laboratories.

In our previous work, we presented MASD [8]. A batch of dry lavender flowers was packed in the MASD system with water. The raw material forms the packed bed. At the bottom, steam is produced by heating water directly with microwave irradiation. Steam produced in the lower part of the apparatus passes through the lavender bed, evaporating and carrying the desired lavender essential oil, and is then directed towards the condenser, located on the top of the main apparatus body. The problem of this system is that all the microwave energy is absorbed by water to heat and vaporise and only a fraction is absorbed by the essential oils inside the lavender flowers.

In this new approach of the improved microwave steam distillation (MSD) (Fig. 1) the important aspect of the extraction reactor is that only the lavender flowers are submitted to the microwave irradiation, resulting in "hot spots" by selective heating. Because essential oil inside lavender flower in general has a significantly higher dielectric loss than the surrounding steam, steams which do not absorb microwaves flow through the lavender flowers which directly absorb microwaves. It is already known that only water in a liquid state absorbs microwaves but steam and ice do not absorb microwaves because in the gas state the molecules are too far from each other to have frictions, and in the solid state the molecules are not free to move and rotate to heat. However, the extraction is conducted in the area of the lavender flower, which is continuously heated by microwaves, resulting in a higher local temperature and hence increased extraction rates.

The aim of this work was to develop and investigate the potential of a new method, MSD, for the extraction of essential oils in

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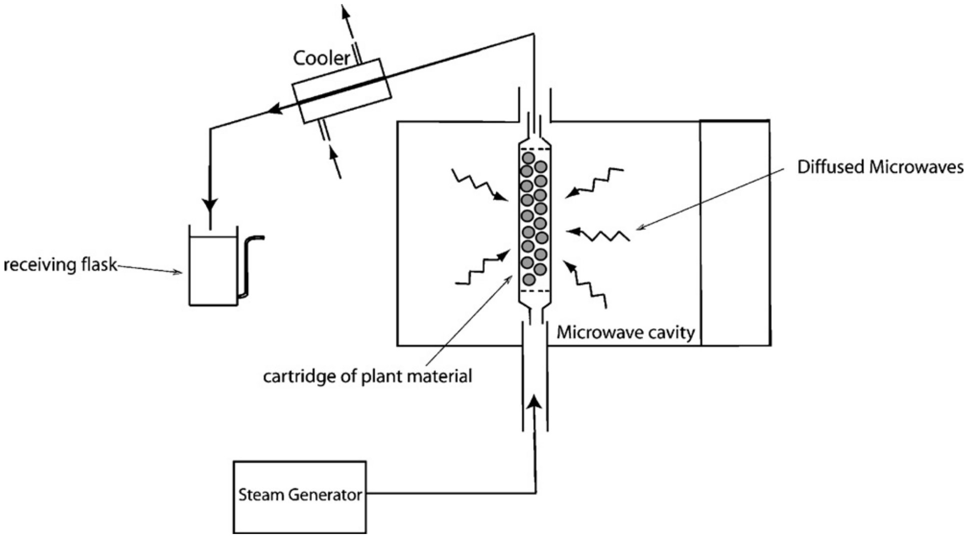

Fig. 1. Improved microwave steam distillation apparatus.

order to introduce this advantageous alternative in the analytical or production of essential oils in food, cosmetic and pharmaceutical industry. Comparisons have been made with conventional steam distillation (SD) for the extraction of essential oils from *Lavandula angustifolia* flowers. We intend to make appropriate comparison in terms of extraction time, yield, aromatic composition, energy used and environmental impact. The chemical analysis studies were supplemented by scanning electron micrographs to shed light on the physical action of the two extraction systems.

2. Experimental

2.1. Plant material

Lavender flowers (*L. angustifolia* Mill., Lamiaceae) were collected in July 2007 in Province area (south France) and let dry on a bench in the shade.

2.2. MSD apparatus and procedure

MSD has been performed in a Milestone “Dry Dist” (Bergamo, Italy) batch reactor. It is a multimode microwave reactor operating at 2.45 GHz with a maximum delivered power of 1000 W variable in 10 W increments. During experiments, time, temperature, pressure and power were controlled with the “easy-WAVE” software package.

An electrical steam generator and a condenser placed outside a microwave zone are connected to a cartridge containing aromatic plant via Pyrex connecting tubes. The condenser is connected to a receiving Florentine flask which is preferably a separating funnel to enable the continuous collection of condensate essential oil and water. This system presents the advantage that the cartridge containing aromatic materials could be easily and quickly replaced and cleaned after each cycle of extraction (Fig. 1).

The cartridge containing 20 g of dry lavender flowers is subjected to microwave heating as soon as the vapour starts to cross the aromatic material. Microwaves distend the plant cells and lead to the rupture of the glands and cell receptacles. The steam passes through the sample, evaporating and carrying the essential oil, and directed towards the condenser and the Florentine flask. The extraction was continued until no more essential oil was obtained. The essential oil is collected, dried with anhydrous sodium sulphate and stored at 4 °C until used. Extractions were performed at least three times, and the mean values were reported.

2.3. Steam distillation apparatus and procedure

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

2.4. Gas chromatography and gas chromatography–mass spectrometry identification

The essential oils were analyzed by gas chromatography (GC) coupled to mass spectrometry (MS) (Hewlett-Packard computerized system comprising a 6890 gas chromatograph coupled to a 5973A mass spectrometer) using two fused silica-capillary columns with different stationary phases. The non-polar column was HP5MS (30 m × 0.25 mm, 0.25-μm film thickness) and the polar one was a Stabilwax consisting of Carbowax-poly(ethylene glycol) (PEG) (60 m × 0.2 mm, 0.25-mm film thickness). GC–MS spectra were obtained using the following conditions: carrier gas He; flow rate 0.3 mL/min; split-less mode; injection volume 1 μL; injection temperature 250 °C; the oven temperature program was 60 °C for 8 min, increased 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 with flame ionization detection (GC–FID). Most constituents were tentatively identified by comparison of their GC Kovats retention indices (*I*), determined with reference to a homologous series of C_5–C_{28} *n*-alkanes and with those of authentic standards available in the authors' laboratory. Identification was confirmed when possible by comparison of their mass spectral fragmentation patterns with those stored in the MS database (US National Institute of Standards and Technology (NIST) and Wiley libraries) and with mass spectra literature data [10–13].

2.5. Scanning electron micrography (SEM)

The specimens were freeze-dried, fixed on the specimen holder with aluminium tape and then sputtered with gold. All the specimens were examined by a Topcon ABT60, under vacuum condition and accelerating voltage of 15 kV, with a spot size of 5 mm and a working distance of 15 mm.

3. Results and discussion

3.1. Preliminary study

In this study, we focused on the potential of MSD which combines the advantages of the steam distillation procedure (extraction of essential oils) and microwave heating (reduction of distillation time). The dielectric constant of steam (water vapour at 100 °C and 1 bar) is very low (1 at 25 °C) in comparison with liquid water (80 at 25 °C) [14]. The important aspect of MSD reactor is that only the lavender flowers are submitted to the microwave irradiation, resulting in "hot spots" by selective heating. Steam is acting only as a carrier gas to make an azeotropic mixture with essential oils and to carry them out of the reactor.

We have optimised steam flow and microwave power with a non-parametric study. A high flow can involve the creation of preferential ways, having consequence less contact between steam and lavender flowers and thus the reduction in essential oil yield. A low flow would be insufficient to extract the totality of essential oil and the extraction time will be too long. The optimum extraction time was obtained for steam flow rate of 8 g/min to ensure the complete extraction yield and the minimum extraction time.

An appropriate microwave irradiation power is important to ensure that the essential oil is extracted quickly; however, the power should not be too high otherwise loss of volatile compounds would result. Different microwave irradiation powers were examined for MSD extraction of essential oils from lavender flowers at the optimum steam flow rate. The total extraction time (it was until no more essential oil was obtained) in relation with the microwave irradiation power was studied. A microwave irradiation power of 200 W for 20 g of lavender flowers was the optimum microwave power density because this power permits in less than 10 min to extract the essential oil completely and to avoid loss of volatile compounds.

3.2. Extraction yield and time

One of the advantages of the MSD method is rapidity. The yields obtained by the two processes are of the same order of magnitude, the only difference observed is the duration of extraction. An extraction time of 6 min for lavender flowers with MSD provides yields comparable to those obtained after 30 min by SD, which is one of the reference methods in essential oil isolation. The values obtained for essential oil yield are 2.7 ± 0.1% for the two processes. Yield is expressed in g of EO per 100 g of dry lavender flowers.

Fig. 2 shows the variation of the extraction yield according to the extraction time. Two phases are observed in the process of the microwave extraction MSD. The first phase is represented by a rapid increase in the yield followed by a second phase which corresponds to a horizontal line which marks the end of the extraction. The rapid increase in the yield during the first step suggests that the essential oil is easily accessible by the steam. Indeed, the microwave radiations distend the plant cells and lead to the rupture of the glands and cell receptacles.

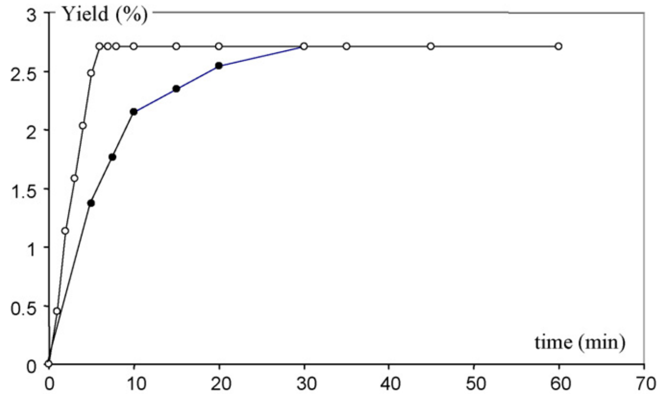

Fig. 2. Yield (g EO/100 g lavender flowers) profile of essential oil obtained by MSD (○) and SD (●) as a function of the extraction time.

Table 1

Chemical composition of lavender EOs obtained by MSD and SD

No.	Compound	I^a	MSD (%)	SD (%)
Monoterpenes				
1	α-Pinene	928	0.67	0.49
2	Camphene	943	0.77	0.60
3	Sabinene	968	0.24	0.18
4	β-Pinene	971	0.79	0.58
5	(*Z*)-β-Ocimene	1035	0.25	0.24
6	(*E*)-β-Ocimene	1045	0.46	0.51
Oxygenated monoterpenes				
7	1,8-Cineole	1033	14.40	13.71
8	Sabinene hydrate-*cis*	1064	0.25	0.27
9	*cis*-Linalool oxide	1069	1.37	1.51
10	*trans*-Linalool oxide	1085	0.66	0.93
11	Linalool	1093	42.52	40.43
12	Camphor	1151	8.11	7.54
13	Citronellal	1150	1.93	2.34
14	Pinocarvone	1163	0.08	0.08
15	Borneol	1176	9.38	10.21
16	Terpinen-4-ol	1179	0.11	0.45
17	α-Terpineol	1189	0.11	0.14
18	Hexyl butyrate	1194	1.14	1.11
19	Verbenone	1200	0.09	0 07
20	Isobornyl acetate	1227	0.33	0.34
21	Carvone	1236	0.32	0.29
22	Linalyl acetate	1259	3.90	4.04
23	Bornyl acetate	1284	0.05	0.05
24	Lavandulyl acetate	1289	0.28	0.31
25	Hexyltiglate	1329	0.60	0.56
26	Geranyl acetate	1377	0.03	0.02
27	Hexyl hexanoate	1383	0.31	0.29
				Sesquiterpenes
27	(*E*)-Caryophyllene	1417	0.47	0.45
28	α-Santalene	1427	0.01	0.01
29	*trans*-α-Bergamotene	1439	0.02	0.01
30	(*E*)-α-Farnesene	1453	0.26	0.26
31	Germacrene-D	1479	0.12	0.14
32	Curcumene	1481	0.01	0.01
Oxygenated sesquiterpenes				
33	Caryophyllene oxide	1589	1.41	1.32
34	α-Cadinol	1651	0.03	0.03
35	α-Bisabolol	1683	0.31	0.36
Other oxygenated compounds				
36	Hexanol	863	0.27	0.23
37	Octan-3-one	983	1.40	1.17
38	Octan-3-ol	996	0.33	0.28
	Extraction time (min)		6	30
	Yield (%)		2.7	2.7

[a] Retention indices relative to C_5–C_{28} *n*-alkanes calculated on non-polar HP5MS capillary column. Percentages calculated by GC–FID on non-polar HP5MS capillary column. Essential oil compounds sorted by chemical families.

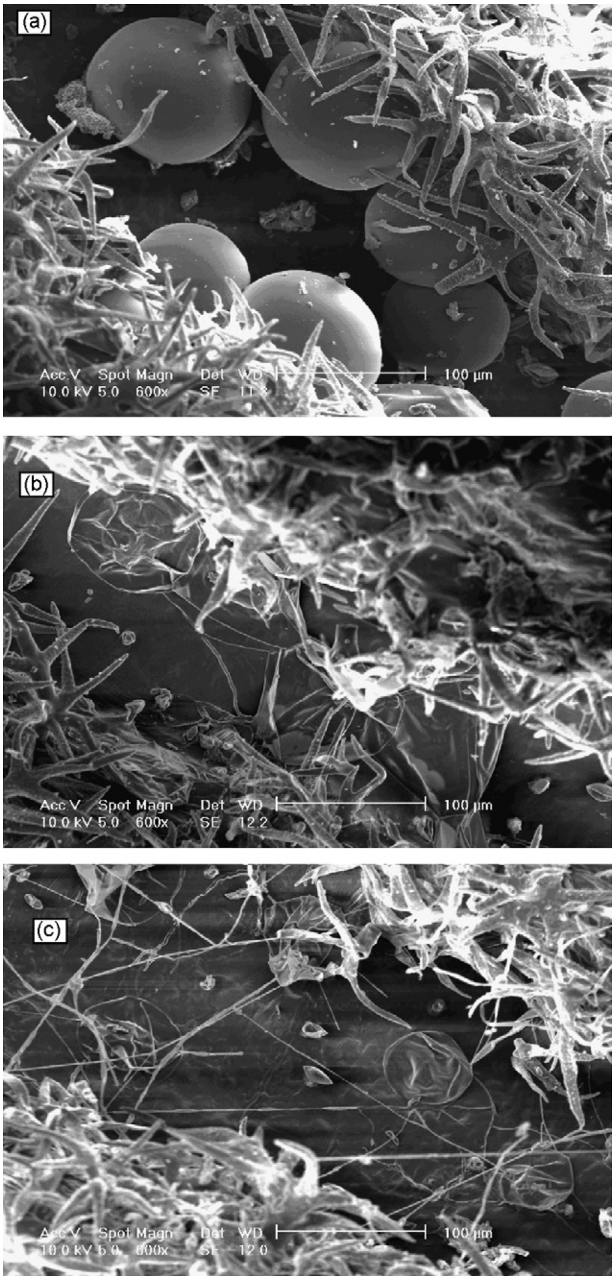

Fig. 3. SEM micrographs of lavender flowers: (a) untreated, (b) after SD isolation (30 min) and (c) after MSD isolation (6 min).

3.3. *Composition of essential oil*

The essential oils of lavender flowers isolated either by MSD or by SD are rather similar in their composition. The same number of volatile secondary metabolites is found in the essential oils with equivalent relative amounts for both extraction methods (Table 1). The essential oil of lavender also contains the same dominant components with comparable contents for MSD and SD: linalool (42.52% and 40.43%), 1,8-cineol (14.40% and 13.71%), borneol (9.38% and 10.21%) and camphor (8.11% and 7.54%). The same conclusion has been reported by Chemat et al. [8] for the essential oil from lavender flowers isolated by MASD and steam distillation. In this application, microwave irradiation highly accelerated the extraction process, but without causing considerable changes in the volatile oil composition, a phenomenon which was already described by Pare and Belanger [15].

3.4. *Structural changes after extraction*

SEM observations on lavender inflorescence calyxes, before oil extraction, confirmed the presence of numerous peltate trichomes with a balloon shape. The glandular trichomes were often hidden by the massive presence of bifurcated non-glandular ones. A generally different feature was shown by glandular trichomes after MSD (Fig. 3). Most of them appeared completely disrupted, particularly after MSD, showing only the remnants of the cuticle. The morpho-structural traits of glandular trichomes, after the different extraction methods, suggest that the main reason for this dramatic reduction in the extraction time is the consequence of trichome cuticle rupture, due to microwave exposure. This rupture has been rarely observed after SD, and thus is conceivable that, during conventional SD, essential oils have to permeate the cuticle to be extracted. This process would require longer times, even though heating the cuticle allows more leakage.

3.5. *Cost, cleanliness and safety considerations*

The reduced cost of steam rather than distillation for essential oil determination in aromatic natural products is clearly advantageous for the proposed MSD method in terms of time and energy. Conventional SD required an extraction time of 30 min. The MSD method required only 6 min for the same amount of essential oil recovery. The energy requirements to perform the distillations are 0.3 kWh/g of EO for SD and 0.05 kWh/g of EO for MSD. The power consumption was determined with a Wattmeter at the microwave generator supply and the steam generator supply. Regarding environmental impact, the calculated quantity of carbon dioxide rejected in the atmosphere is higher in the case of SD (240 g CO_2/g of EO) than of MSD (40 g CO_2/g of EO). These calculations have been made based on the following assumptions: to obtain 1 kWh from coal or fuel, 800 g of CO_2 will be rejected in the atmosphere during combustion of fossil fuel [16].

4. Conclusion

In this study, we propose a new process of the extraction of essential oil: MSD. It is an original combination of microwave heating and steam distillation. The efficiency of the new technique MSD is considerably higher than the conventional procedure SD if we take into account short distillation times required, cost and energy used and cleanliness of the process. MSD highly accelerated the isolation process, without causing changes in the volatile oil composition. This MSD method could be appropriate for the routine quality control analysis of essential oils from aromatic herbs, spices or flowers.

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