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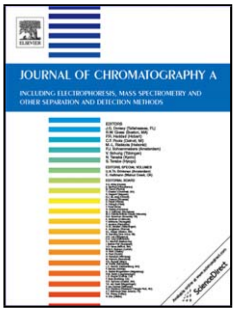

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Eco-friendly and cleaner process for isolation of essential oil using microwave energy.

Experimental and theoretical study

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Abstract

Microwave steam diffusion (MSD) was developed as a cleaner and new process design and operation for isolation of essentials oils and was compared to conventional steam diffusion (SD). The essential oils extracted by MSD for 3 min were quantitatively (yield) and qualitatively (aromatic profile) similar to those obtained by conventional steam diffusion for 20 min. In addition, an optimal operating steam flow rate of 25 g min^{-1} and microwave power 200 W were found to ensure complete extraction yield with reduced extraction time. To confirm the efficiency of this process a mathematical model was proposed to describe the mass transfer of essential oil from lavender. Solid-steam mass transfer coefficients obtained by MSD was six times higher than obtained by SD. Scanning electronic microscopy was used to confirm the extraction mechanism of the essential oil present in the glandular trichomes of the flowers from lavender outer surface. MSD was better than SD in terms of energy saving, cleanliness and reduced waste water.

Keywords**:** Microwave extraction, Essential oil, Lavender, steam diffusion, Green analysis.

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1. Introduction

Microwave energy is well known to have a significant effect on the rate of various processes in the chemical and food industry. Much attention has been given to the application of microwave dielectric heating for the extraction of natural products that typically needed hours or days to reach completion with conventional methods. Using microwaves, full reproducible extractions can now be completed in seconds or minutes with high reproducibility, reducing the consumption of solvent, simplifying manipulation and work-up, giving higher purity of the final product, eliminating post-treatment of waste water and consuming only a fraction of the energy normally needed for a conventional extraction method such as steam distillation or solvent extraction. Several classes of compounds such as essential oils, aromas, pigments, anti-oxidants, and other organic compounds have been extracted efficiently from a variety of matrices (mainly animal tissues, food, and plant materials). Microwave extraction is a research area which has an impact in several fields of modern chemistry. All the reported applications have shown that microwave assisted extraction is an alternative to conventional techniques for such matrices. The main benefits are decrease of extraction time and solvents used. The advantages of using microwave energy, which is a non contact heat source, for the extraction of essential oils from plant materials, includes: more effective heating, faster energy transfer, reduced thermal gradients, selective heating, reduced equipment size, faster response to process heating control, faster start-up, increased production, and elimination of process steps. Extraction processes performed under the action of microwave radiation are believed to be affected in part by polarization, volumetric and selective heating [1].

Driven by reducing energy and waste water or solvent, advances in microwave extraction have resulted in a number of techniques such as microwave-assisted solvent extraction (MASE) [2-3], compressed air microwave distillation (CAMD) [4], vacuum microwave hydrodistillation (VMHD) [5], microwave hydrodistillation (MWHD) [6], solvent free microwave extraction (SFME) [7], microwave-accelerated steam distillation (MASD) [8-9] and microwave hydrodiffusion and gravity (MHG) [10]. Specifically in the essential oil extraction, microwave mediated processes are highly desirable due to their small equipment size, simple, rapid, controllability through mild increments of heating and low solvent consumption, thus reducing environmental impact and costs.

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This present study has been planned with the aim to design, optimize and operate a new and green process microwave steam diffusion (MSD) (Fig. 1) for the extraction of essential oil from lavender. There have been two reported articles in the literature regarding 'microwave' extraction techniques of lavender essential oils. Chemat et al. [8,9] reported a new process design and operation for microwave accelerated steam distillation (MASD). A batch of dry lavender flowers was packed in the MASD 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. Also, Chemat et al. [11] studied another type of new microwave design: microwave-assisted hydrodistillation (MAHD). In this case flowers were soaked in distilled water before microwave processing. Certainly, the most remarkable benefit of the both process (MAHD and MASD) is its rapidity and, energy saving. However, the most problem of these systems 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.

MSD was designed and constructed that only the lavender flowers are submitted to the microwave irradiation, resulting in "hot spots" by selective heating. 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 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. On the other hand, this green method used the downward saturated steam, allows the extract to diffuse outside the plant material and drop by earth gravity out the microwave reactor.

In order to obtain a better understanding of microwave steam diffusion, comparisons have been made, in term of extraction time, yield, mass transfer coefficients from solid to liquid phases, aromatic composition, energy used and environmental impact with conventional steam diffusion (SD). The chemical analysis studies were supplemented by scanning electron micrographs to shed light on the physical action of the two extraction systems.

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2. Experimental

2.1. *Plant material*

Lavender flowers (*Lavandula angustifolia* Mill., *Lamiaceae,* Lamiaceae) were collected in July 2007 from a private garden in Avignon province (south France) and allowed to dry on a bench in the shade.

2.2. *Gas chromatography-mass spectrometry*

Essential oil composition was determined by gas chromatography coupled to mass spectrometry (GC–MS) analysis on a Hewlett-Packard 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 x 0.25 mm, 0.25 µm film thickness) and the polar column was a Stabilwax consisting (60 m x 0.25 mm, 0.25 µm film thickness). GC–MS spectra were obtained using the following conditions: the vector gas was helium, the flow rate for the HP-5MS column was 0.3 ml/min and with the Stabilwax column was 0.7 ml/min, mode, splitless injection volume, 1 µl; injection temperature, 250 °C; oven temperature programme, 60 °C for 8 min, then increased at 2 °C/min to 250 °C and held at 250 °C for 15 min; ionization mode, electronic impact at 70 eV.

Identification of the components was based on computer matching against commercial libraries (Wiley, MassFinder 2.1 Library, and NIST98), laboratory mass spectra libraries built up from pure substances, and MS literature data combined with the comparison of GC retention indices (*I*) on apolar and polar column. *I* values were calculated with the help of a series of linear alkanes C_8–C_{26} on apolar and polar column (HP5MS and Stabilwax). Compounds available in the laboratory were confirmed by external standard compound co-injection. Each sample was analyzed three times in both columns.

2.3. *Scanning electron microscopy (SEM)*

Microhistological analysis was done by using a FEIXL30 scanning electron microscope equipped with a Centaurus detector for backscattered electron image acquisition. Single flowers were individually packed in pieces of perforated aluminium foil to permit gas exchange, rapidly frozen in liquid nitrogen before being transferred to a pre-cooled (−60 °C) plate of a Pearse tissue dryer and dried overnight under vacuum. Samples were then coated with a thin gold film.

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2.4. Microwave steam diffusion apparatus and procedure

MSD has been designed and constructed in a Milestone microwave laboratory oven [12] showed in Fig. 1. This is a multimode reactor 2.45 GHz with a maximum delivered power of 1000 W. Time, temperature, pressure and power were controlled with the "easy WAVE" software package during experiments.

A batch of 30 g lavender flowers was packed in 1.5 litre reactor at a time. An electrical steam generator and a conventional condenser placed outside a microwave zone are connected to a reactor containing the raw material, forms the packed bed, via Pyrex connecting tubes (10 cm). The condenser is connected to a receiving Florentine flask which is preferably a separating funnel to enabling the continuously collected of condensate essential oil and water. This system presents the advantage that the reactor containing aromatic particles could be easily and quickly replaced and cleaned after each cycle of extraction.

MSD is a new process design involves removal of essential oil from the raw material forms the packed bed into the downward saturated steam, while the mixture is continuously heated in a microwave cavity. The direct interaction of microwaves with saturated steam favours the release of essential oils trapped inside the cells of plant tissues. A mixture of hot "crude juice" and steam move thus naturally downwards by earth gravity downwards, and directed towards the condenser to 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. A wattmeter (W) has been added at the generator entrance, in order to measure the power consumption. Extractions were performed at least three times, and the mean values were reported.

2.5. Steam diffusion apparatus and procedure

For a rigorous comparison, the same glassware and same operating conditions have been used for conventional steam diffusion (the same process but without use of microwave cavity). In this system, the vapour produced by the steam generator crosses the plant, charged with essential oil then pass through the condenser to a receiving 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. A wattmeter (W) has been added at the generator entrance, in order to measure the power consumption. Extractions were performed at least three times, and the mean values were reported.

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3. Results and discussion

3.1 Optimization of MSD parameters

The validation of the proposed method has been performed for determination of lavender essential oils. The main parameters that affect the extraction efficiency of MSD are steam mass flow rate (G_v) and the microwave power (P_w). The optimization of the experimental parameters can be carried out step-by-step. Table 1 shows different conditions of experiments carried out with the MSD for extractions of lavender. All of the selected factors were examined by using five-level.

3.1.1 Effect of steam mass flow rate

Extraction rate increases with steam flow because of the fact that steam in extraction reactor has two uses: to provide heat energy and as a transport medium for the distillate. A low flow would be insufficient to extract the totality of essential oil and the extraction time will be too long. Extraction rate of lavender oil extractions were very faster at higher flow rates. It means that the mass transfer of lavender oil from the surface of the solid phase into the steam phase regulated most of the extraction process. Increase of flow rate resulted in increase of superficial velocity and thus quicker mass transfer (Fig. 3).

In order to understanding how the optimum of steam flow was obtained from 25 g min^{1}. Thus, the first order kinetics was proposed to explain the extraction essential of lavender. The kinetics was found to be well fitted with an equation of the form:

$$\mathrm{Ln}\left(\frac{Y\ -\ Y(t)}{Y}\right) = -\,k_c \bullet t \quad \textbf{(1)}$$

where

r =yield of lavender essential oil in each time (%)

$r(t)$=overall yield of lavender essential oil in the end of extraction (%)

k_c= constant kinetic (min^{-1})

We draw the curve corresponding to kinetic constant K_c in function of steam flow rate. Fig.4 shows that from 25 g min^{-1} the constant kinetic K_c was stable. This could be the consequence of the constant pressure and, in turn, the constant steam temperature, and would explain the minimum extraction time obtained with this method from 25 g min^{-1}.

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Thus, from this value of steam flow the temperature brought by the steam phase into the surface of the solid phase was stable. The optimum extraction time was obtained from steam flow rate equal to 25 g min^{-1} to ensure complete extraction yield and the minimum extraction time.

3.1.1 Effect of microwave power

Data presented in Fig.5 show the variations in the extraction yield as a function of the microwave power. No striking differences were found in oil yield in different levels. An appropriate microwave irradiation power is important to ensure 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 of lavender flowers was the optimum microwave power density because this power permits in less than 3 min to extract the essential oil completely and avoid loss of volatile compounds.

The first order kinetics was proposed to explain the rate of extraction of essential oil from lavender. We draw the curve corresponding which the value of the constant kinetic K_c presented in Fig.6 show that at MW power higher than 200 W the constant kinetic K_c was stable. This could be the consequence of the increase of the temperature inside the sample, leading to the rupture of the essential oil cells, followed by evaporation of the water and essential oils. The power must be sufficient to reach the boiling point of the water (100 °C), which fixes the steam temperature. However, the power should not be too high (at 300 W and 400 W), otherwise loss of volatile compounds would result.

The both experimental MSD variables were optimized in order to maximize the yield and reduce extraction time. The highest extraction efficiency was obtained under these optimal conditions: steam flow rate $G_v = 25$ g min^{-1} and incident microwave power $P_w = 200$ W.

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3.2 Composition of essential oil

The retention time, molecular formula, content, and total content of chemical compositions in essential oils from dried lavender obtained by two kinds of extraction methods at the optimal conditions, were listed in Tables 2. The essential oils of lavender flowers extracted by MSD or SD are rather similar in their composition. The same number of volatile secondary metabolites is found in the essential oils isolated by MSD or SD, with similar yields. The essential oil of lavender also contains the same dominant components with comparable contents for MSD and SD, respectively. In this application, microwave irradiation highly accelerated the extraction process, but without causing considerable changes in the volatile oil composition, phenomenon which was already described by Pare and Belanger [18].

3.3 Microhistology analysis

The literature revealed that lavender glandular trichomes appeared swollen, with a highly wrinkled surface and empty of essential oil, but the integrity of their cuticles was maintained [11,16]. In this paper, the various extraction methods produced distinguishable physical changes in the lavender calyxes. Fig. 10a is a micrograph of the untreated flowers, which can be compared with structures of the treated lavender calyxes in Fig. 10b (SD) and Fig. 10c (MSD). Fig. 10a shows the typical structure before oil extraction; the SEM images reveal the presence of numerous peltate trichomes with a balloon shape and a smooth head, mainly distributed along the outer surface of flowers calyx ribs. The glandular trichomes were often hidden by the massive presence of bifurcated non glandular ones. After SD, all peltate trichomes reduced their volume, empty of essential oil, with a highly wrinkled surface (Fig.10b). In the case of MSD extraction, a generally different feature was shown by glandular trichomes (Fig.10c). Most of them appeared completely disrupted, particularly after MSD, showing only remnants of the cuticle. Thus, in the case of microwave heating, the glandular trichomes were subjected to more severe thermal stresses and localized high pressures; then this pressure build-up within the glandular could have exceeded their capacity for expansion, and caused their rupture more rapidly than in SD extraction.

Similar effects were pointed out by Iriti et al. [11] for the different variants of the microwave-assisted hydrodistillation technique (MADH) of lavender flowers. When the morpho-structural traits of glandular trichomes were suggest that the main reason for this dramatic reduction in the extraction time is trichome cuticle rupture, due to microwave exposure.

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3.4 Extraction kinetics and modelling

3.4.1 Extraction kinetics

From data obtained from extraction experiments with two processes, a curve of the overall yield of essential oils content in the flowers as function of time was plotted, at optimal operating conditions. No differences were found in oil yield obtained by the two techniques. As is shown in Fig. 4, an extraction time of 3 min with MSD provides yields comparable to those obtained after 20 min by means of SD. The overall yield of essential oils obtained from lavender flowers was 4.42% and 4.43% by MSD and SD, respectively. Thus, one of the advantages of the MSD method is rapidity.

Besides, Fig.4 shows, for two techniques, the process of the kinetics of extraction is performed in two distinctive parts:

(i) Part 1 of the curve corresponding to a rapid increase in the yield, suggests that the essential oil is easily accessible by the steam. It is also the phase of extraction of essential oil in flowers.

(ii) Part 2 of the curve represents a horizontal line which marks the end of the extraction.

Thus, it was found that the change between the two phases is partly due to changes in the mechanisms of mass transfer and partly due to changes in the surface area from which oil evaporates. In this respect and in order to obtain a better understanding of lavender essential oil obtained by MSD a solid-steam mathematical model was proposed to describe the mass transfer in the oil obtaining process.

3.4.2 Modeling

The aim of part of this work was to explain how microwave steam diffusion (MSD) highly accelerated the extraction process, without causing considerable changes in the volatile oil composition and properties. Thus, to explain the microwave steam diffusion of lavender essential oil, a mathematical model is proposed by Romdhane et al. [13]. However, in this paper, this model was applied with some modification.

All authors have attempted to model the kinetics of steam oil extraction processes considers the composition of the oil as being made up of only the single component [13, 14, 15]. However, it is important to note that one unique study attempted to understanding the lavandin super steam distillation using a phenomenological model by considering three series stages in the oil obtaining process [16]. Thus, in this paper, the necessity of implementation of

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different volatilities for essential oil components has been suggested. Therefore, the lavander essential oil is considered as a mixture of the two major components (ie linalool and linalyl acetate) and to keep the same following assumptions have been made. The model and the experimental data were used to determine the mass transfer parameters including an overall mass transfer coefficient.

3.4.2.a Mass balance:

The overall oil mass balance made over the column boundaries gives:

$$G_v\, y_0 - G_v\, y(t) = \frac{dm}{dt} \qquad \mathbf{(2)}$$

where

m =is the mass of oil contained in the column,

y_0=is the input essential oil fraction by weight,

y(t) =is the steam essential oil fraction by mass in the outlet of the column at a time t,

G_v = is the mass flow rate of steam (g min^{-1}).

Consider that M_i is the mass of the inert part of the flowers (excluding the essential oil) and *M* is the mass of the plant material. At any time t the relationship between M_i and M is given by:

$$M_i = M\,(1 - x\,(t)) \qquad \mathbf{(3)}$$

where *x (t)* is the fraction by mass of oil in the seeds at a time t.

The mass of oil in the column at a time t is then given by:

$$m(t) = M\,x\,(t) + \varepsilon\,\rho_s\,V_c\,y(t) \qquad \mathbf{(4)}$$

The following equation gives *x(t)*.

$$x(t) = 1 - \frac{1 - x_0}{1 - \frac{G_v}{M_0}\int_0^t y(t)\,dt + \frac{\varepsilon\,\rho_s\,V_c}{M_0}\left(y_0 - y\right)} \qquad \mathbf{(5)}$$

3.4.2.b Rate of extraction

The rate at which oil is transferred to steam can be described in terms of an overall mass transfer coefficient *K* and the difference between an average composition of the seeds, *x*, and an equilibrium composition, *x**, Eq (6).

$$-\frac{dm}{dt} = K\,\Omega\,\left(x - x^*\right) \qquad \mathbf{(6)}$$

where

K =is an overall mass transfer coefficient (g oil min^{-1} m^{-2}),

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Ω =is the surface area available for mass transfer (m^2),

x^* =is an oil fraction in the flowers assumed in equilibrium with steam.

It is difficult to determine x^* directly; therefore it is proposed to assume that x^* is proportional to the oil vapour pressure at a given temperature. The essential oil of lavender was considered as a mixture of his two major components, therefore the oil vapour pressure is assumed to equal that of linalool and linalyl acetate:

$$x^* = \sum_{i=1}^{2} \alpha P^0 \quad \mathbf{(7)}$$

where

α =is proportionality parameter,

P^o =is the pure vapour pressure of linalool and linalyl acetate at a temperature T.

Consider specific mass transfer area, a ($m^2\ kg^{-1}$) defined by $a = \Omega / M_0$ and an in-column steam to material ratio, $\check{s}$ defined by $\check{s} = \varepsilon\rho_s V_C/M_0$, Equation (8) can be rewritten simply as follows:

$$\frac{1 - x_0}{(1 - x)^2}\frac{dx}{dt} + \bar{s}\frac{dy}{dt} = -Kax + \alpha K a P^0 \quad \mathbf{(8)}$$

Values for Ka and α were determined by fitting the experimental results to Eq (8). Indeed, graphical representation of the left-hand side of Eq (8) as function of x would result in a line of slope −Ka and intercept $\alpha K a P^o$. The variation of P^o was determined by computer programs namely ACDLabs 7.0.

3.4.2.c Determination of the mass transfer coefficient and the proportionality parameter α

The mass transfer coefficient, *K*, at optimal operating conditions was determined using the model developed earlier (Eq. 8) combined with experimental data. The mass transfer coefficient *K* is determined if the specific area, *a*, is known. The lavender particles were assumed to be spherical and their average diameter was determined using the sieving method; a value of 2mm was obtained. The specific mass transfer area, *a*, is then calculated to be 0,0158 $m^2\ g^{-1}$. Values for *K* and *α* were determined for the two processes and were showed in Table 3. The overall mass transfer coefficient *K* obtained by MSD was six times bigger than obtained by SD. These results explain and confirm the fact of the internal microwave heating of the steam within the solid samples (lavender flowers) facilitates the diffusion of glands and free bio-molecules (essential oil) from the inside of the flowers to the steam by physical phenomenon, namely hydrodiffusion.

3.4.2.d Rate of extraction

From data obtained from extraction experiments by MSD and SD, a curve of the oil content in the flowers as function of time was plotted (Fig. 4). The data were converted to rates of extraction, expressed as *N* (g oil extracted $\text{min}^{-1}\text{m}^{-2}$), and plotted against flowers oil content, as shown in Fig 9. This was done by measuring small changes in oil content, (Δx) for corresponding small changes in time (Δt), and calculating the rate as:

$$N = \frac{-M_i \Delta x}{\Omega \Delta t} \qquad \mathbf{(9)}$$

Therefore, as is shown in Fig. 9, MSD is clearly quicker than conventional SD and confirm the results obtained in Fig. 7. It was found that the mass transfer has increased by almost six times (Table 3) when the microwave power has applied at optimal conditions. This is explained by the fact that an increase in the two transfer phenomena mass and heat (Fig. 8) are in the same direction from the inside to the outside of the flowers, which facilitates oil diffusion from the inside of the flowers to steam by increasing of temperature results in an application of microwave power. In addition it was mentioned in the SEM study that the structure of the flowers was modified following an application of microwave power. Thus, the morpho-structural traits of glandular trichomes, after the microwave extraction, suggest that the main reason for this dramatic reduction in the extraction time is the consequence of trichome cuticle rupture, due to microwave exposure and results in mass transfer increase. Therefore, the extraction of essential oils from lavender with MSD was better than SD in terms of energy saving, cleanliness and product quality.

3.5 Cost, energy, and environment ecology

The reduced cost of isolation is clearly advantageous for the proposed MSD method in terms of energy and time. The power consumption has been determined with a Wattmeter at the microwave generator entrance and the electrical heater power supply. Regarding environmental impact, the calculated quantity of carbon dioxide rejected in the atmosphere is higher in the case of SD (338 kg CO_2 kg $^{-1}$ oil) than for MSD (69.3 kg CO_2 kg $^{-1}$oil) (see Table 4). These calculations have been made according to the literature: to obtain 1 kWh from coal or fuel, 800 g of CO_2 will be rejected in the atmosphere during combustion of fossil fuel [17]. Waste water was also reduced from 413 liter kg^{-1} EO with SD to 62 liter kg^{-1} EO with MSD.

4. Conclusion

An innovative and cleaner design process for extraction of essential oil was developed in this paper: MSD. This green process has been studied and tested using dry lavender flowers. This new system was developed to date indicate that microwave extraction process offers important advantages over traditional steam diffusion, namely, shorter extraction times (3 min against 20 min), less energy consuming and lower cost.

The general goal of this work was to confirm the efficiency of this new microwave system and to explain how microwave extraction highly accelerated the extraction process, without causing considerable changes in the volatile oil composition and properties. Therefore, a mathematical model was proposed to describe the mass transfer of essential oil extracted by MSD. Two model parameters were calculated by experimental data correlation for both methods (MSD and SD): overall mass transfer coefficient ($K_{MSD} = 32.230$ g m^{-2} min^{-1}, $K_{SD} = 6.375$ g m^{-2} min^{-1}) and proportionality parameter ($\alpha_{MSD} = 0.387$ bar^{-1}, $\alpha_{SD} = 0.186$ bar^{-1}). These parameters shown that with MSD the mass transfer coefficient was six times bigger than obtained by SD. In addition, the SEM was used to confirm how the essential oil present in the glandular trichomes of lavender flowers diffused outside the plant material. The result shown that after MSD and SD, all observed glandular trichomes reduced their volume and looked like imploded, but maintaining the integrality of their cuticles. Therefore, the performance of the microwave steam diffusion was studied with a univariate study in order to quantify the variables which may minimize the time of extraction of essential oil from lavender flowers. The two variables chosen, namely steam flow rate and microwave power. The highest extraction efficiency was obtained under these conditions: $G_v = 25$ g min^{-1} and $P_w = 200$ W. Finally, in this study, we present microwave steam diffusion as a green, cleaner, an environmentally friendly and economic procedure.

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Notation

A = Specific interfacial area per unit mass of bed ($m^2\ kg^{-1}$)

G_v = Mass flow rate of steam (g min^{-1})

K = Overall mass transfer coefficient (g oil $min^{-1}m^{-2}$)

K_c = Kinetic constant (min^{-1})

M =Mass of the flowers (g)

m = Mass of oil contained in the flowers (g)

M_i = Mass of the inert part of the flowers (excluding the essential oil) (g)

N = Rate of extraction (g $min^{-1}\ m^{-2}$)

P = Pressure (bar)

P^o =Tension vapour (bar)

P_w = Microwave power (W)

$\check{S}$ = In-column steam to material ratio (g steam g^{-1} material)

t = Time (min)

T = Temperature (K)

V_C = Column volume (m^3)

x = Flowers oil fraction by mass (g essential oil g^{-1} flowers)

x_0 = Critical oil content (g essential oil g^{-1} flowers)

y =Steam oil fraction by mass (g essential oil g^{-1} steam)

Y =Extraction yield (%, g g^{-1})

α =Proportionality parameter (bar^{-1})

ε =Column voidage

ρ_s =Steam density (kg m^{-3})

Ω =Surface area available for mass transfer (m^2)

Superscripts

0 =Initial

f =Final

in =Input

T =Total

Subscripts

$*$ = Equilibrium

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Figure 1. Schematic diagram of the Microwave steam diffusion process.

Figure 2. Conventional steam diffusion (SD).

Figure 3. Yield profile of essential oil from lavender as function of the mass flow rate of steam.

Figure 4. Kinetic constant and the infinite time of extraction of essential oil from lavender as function of the mass flow rate of steam.

Figure 5. Yield profile of essential oil from lavender as function of the microwave power.

Figure 6. Kinetic constant and the infinite time of extraction of essential oil from lavender as function of the microwave power.

Figure 7. Yield profile of lavender essential oil obtained by MSD (■) and SD (□) as function of the extraction time.

Figure 8. Oil transfer route from the trichome to the steam flow bluk.

Figure 9. Rate of extraction of lavender oil (g min^{-1} m^{-2}) as function of lavender oil fraction by mass (g oil g^{-1} solid) obtained by MSD (■) and SD (□).

Figure 10. SEM micrographs showing the morphology of flower calyx trichomes of lavender, before (a), after SD extraction (b) and after MSD extraction (c).

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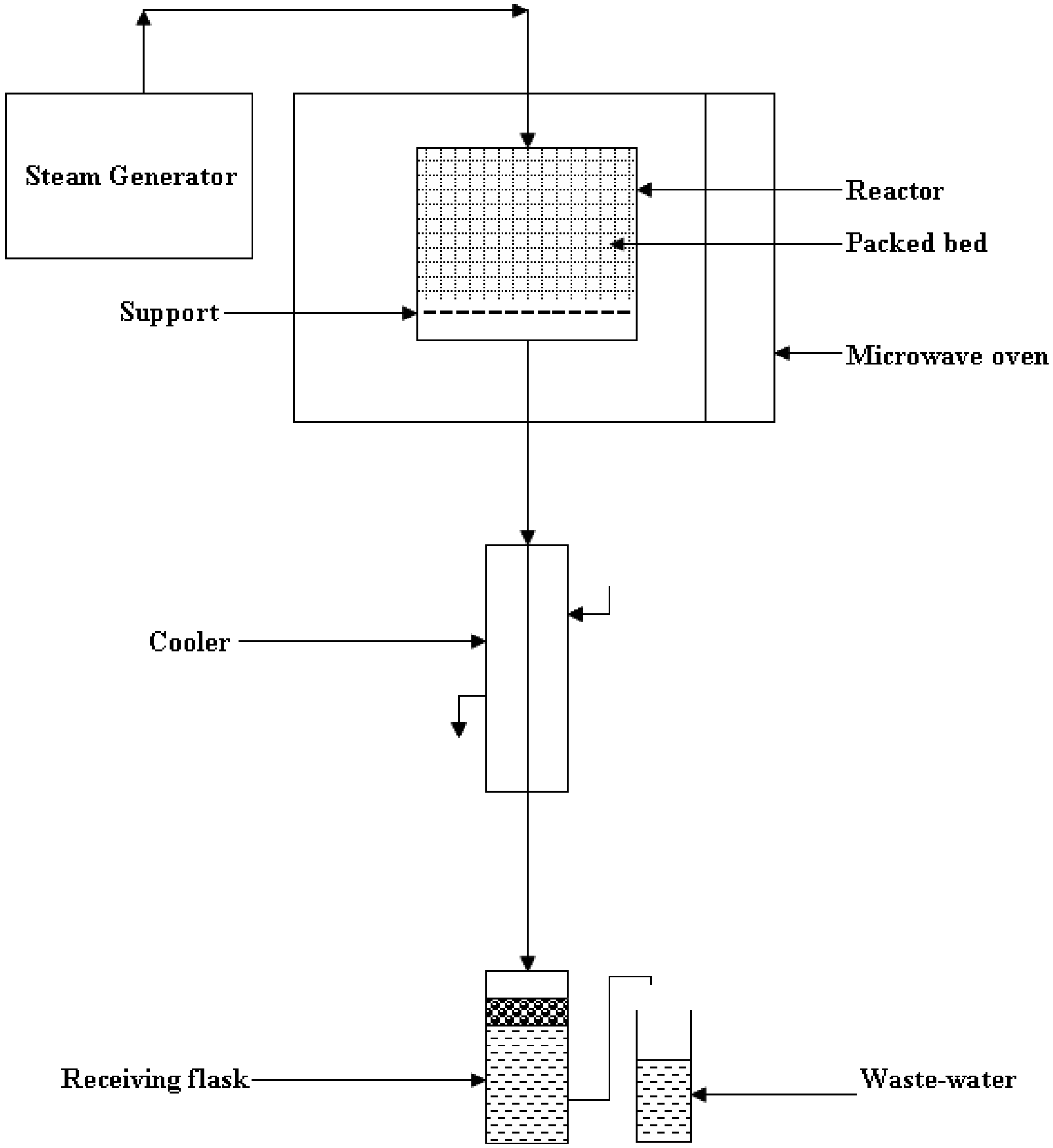
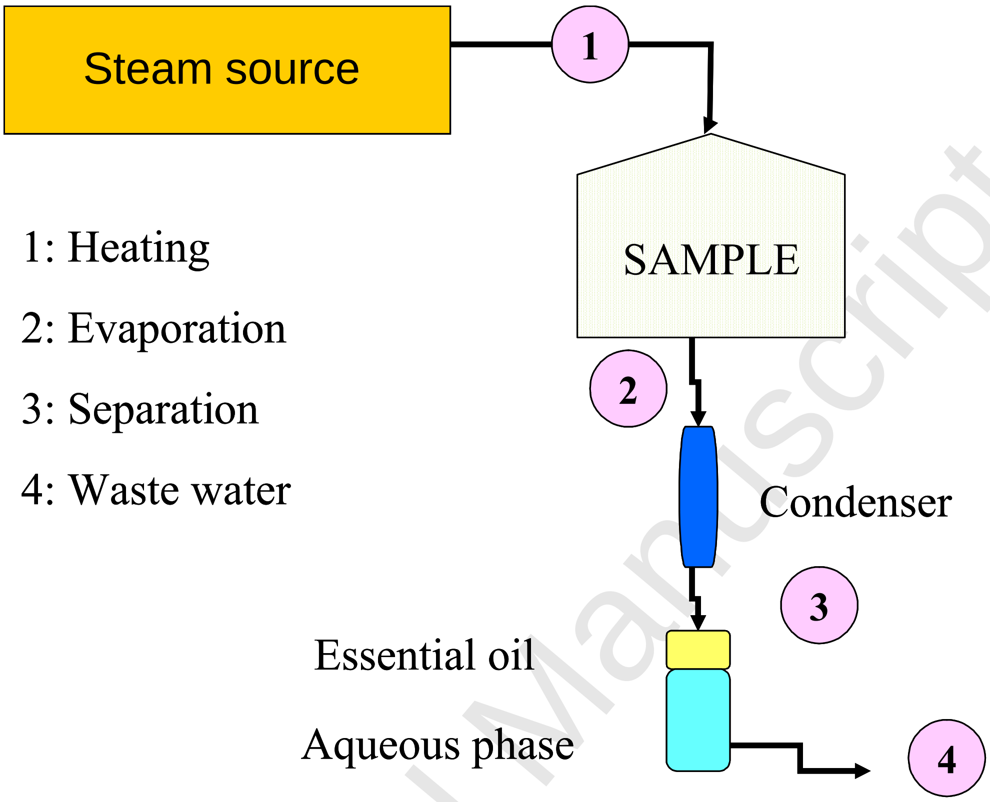

Steam Generator
Reactor
Packed bed
Support
Microwave oven
Cooler
Receiving flask
Waste-water


Steam source
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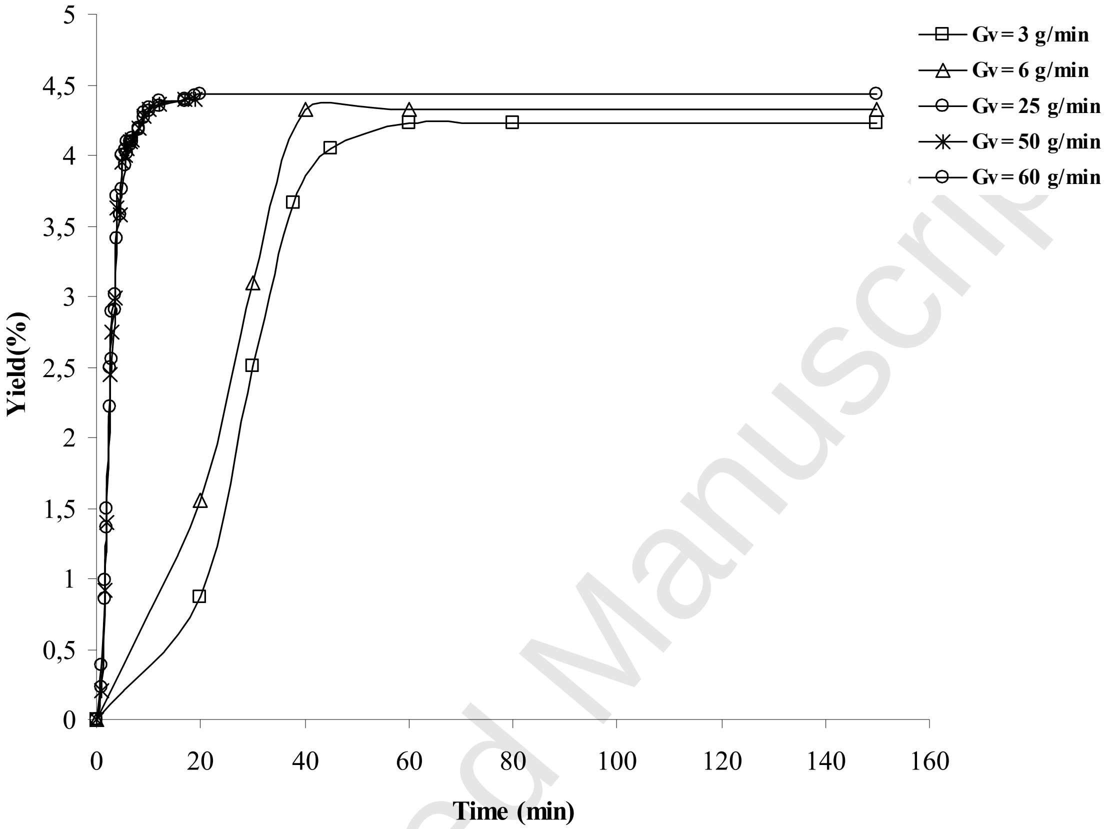
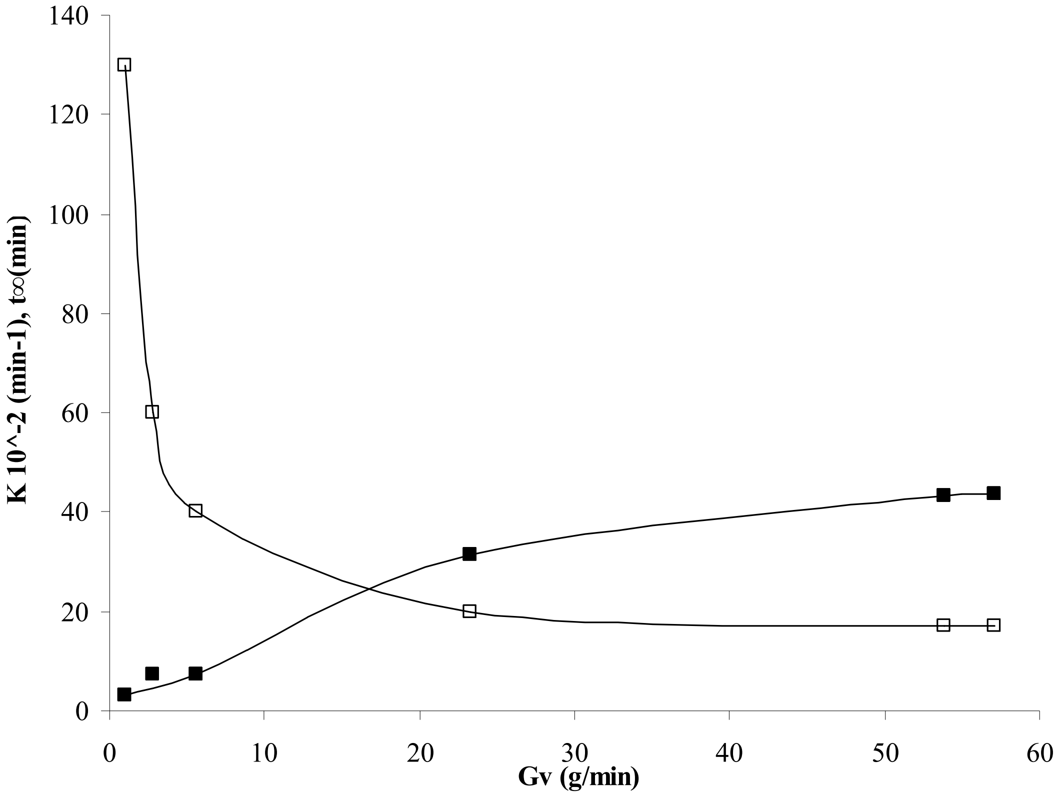
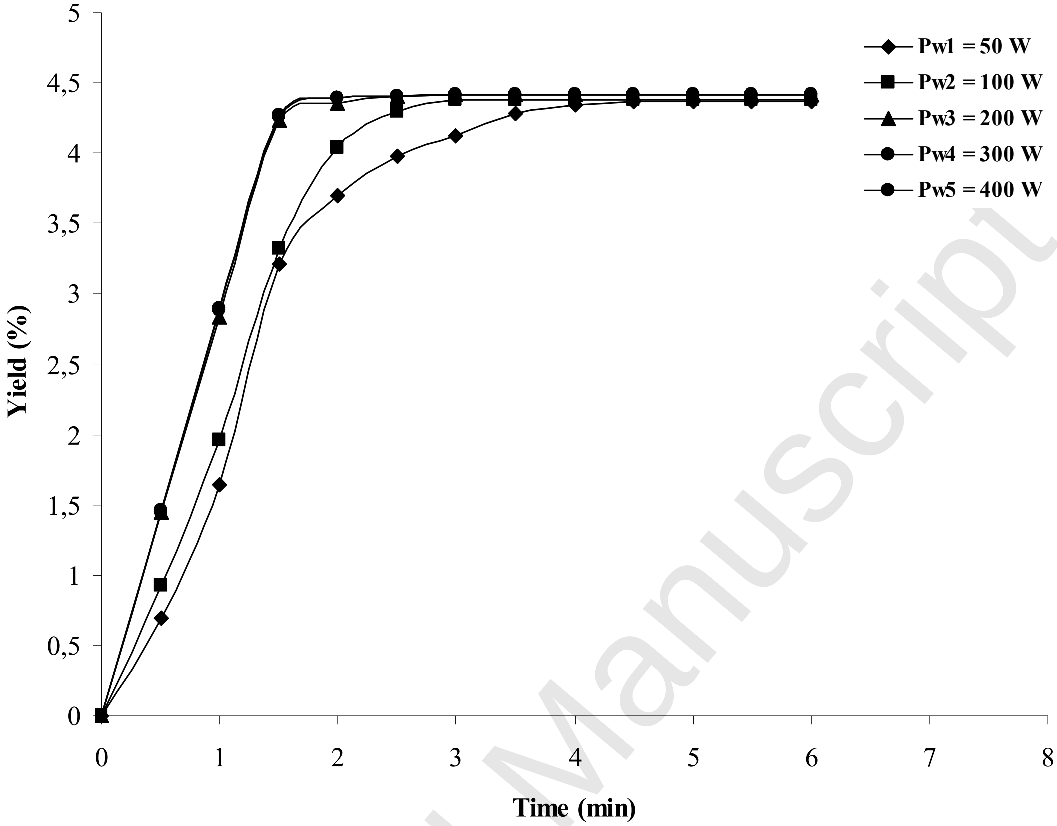
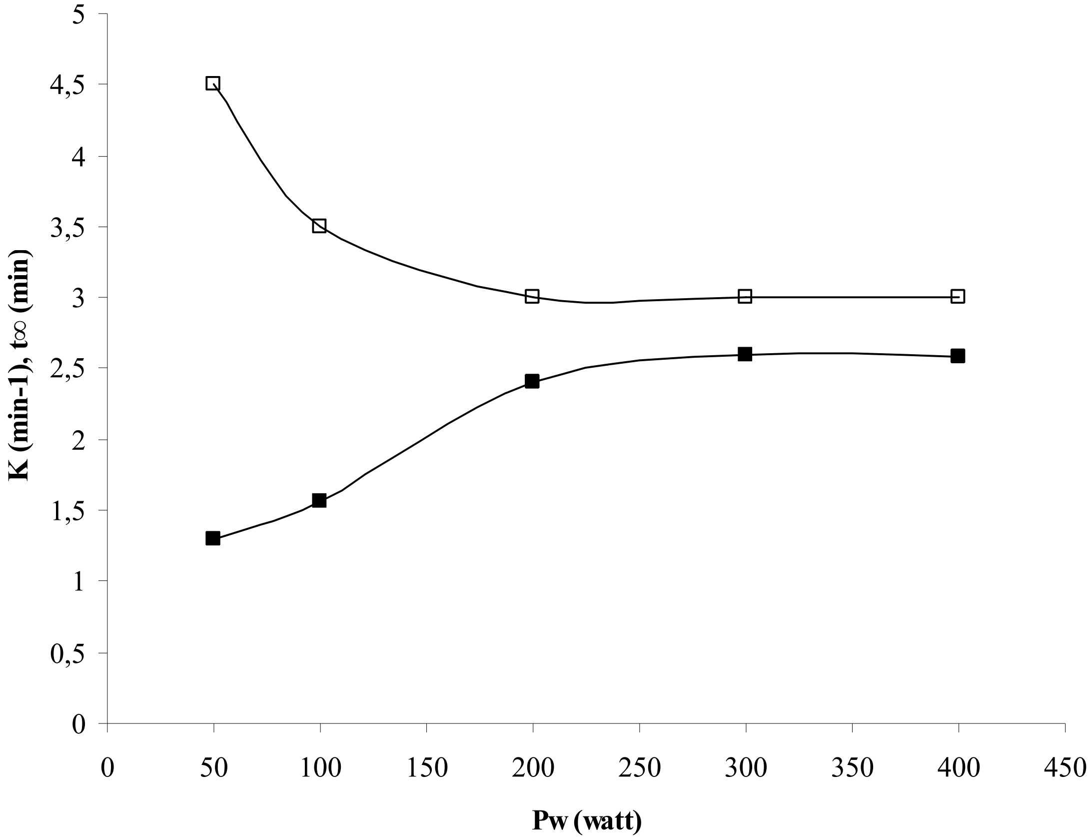
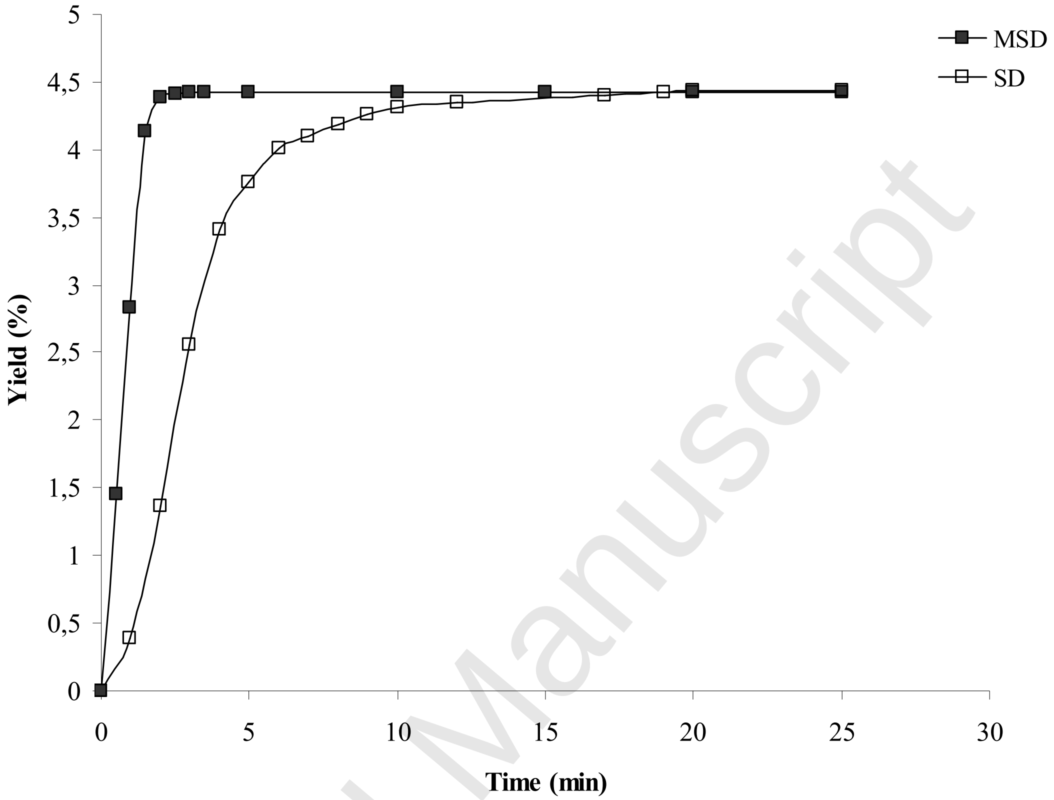
SAMPLE
1: Heating
2: Evaporation
3: Separation
4: Waste water
2
Condenser
3
Essential oil
Aqueous phase
4


5
4,5
4
3,5
3
2,5
2
1,5
1
0,5
0
Yield(%)
0
20
40
60
80
100
120
140
160
Time (min)
Gv = 3 g/min
Gv = 6 g/min
Gv = 25 g/min
Gv = 50 g/min
Gv = 60 g/min


140
120
100
80
60
40
20
0
K 10^-2 (min-1), t∞(min)
0
10
20
30
40
50
60
Gv (g/min)


5
4,5
4
3,5
3
2,5
2
1,5
1
0,5
0
Yield (%)
0
1
2
3
4
5
6
7
8
Time (min)
Pw1 = 50 W
Pw2 = 100 W
Pw3 = 200 W
Pw4 = 300 W
Pw5 = 400 W


5
4,5
4
3,5
3
2,5
2
1,5
1
0,5
0
K (min-1), t∞ (min)
0
50
100
150
200
250
300
350
400
450
Pw (watt)



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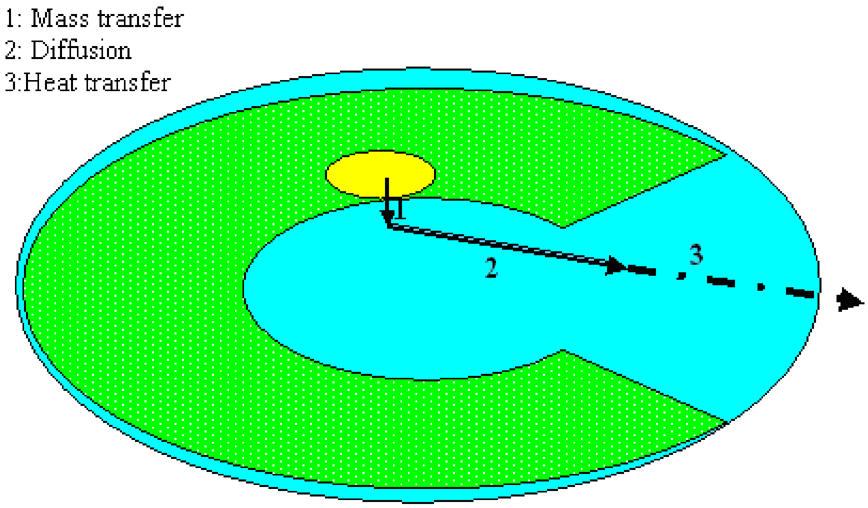

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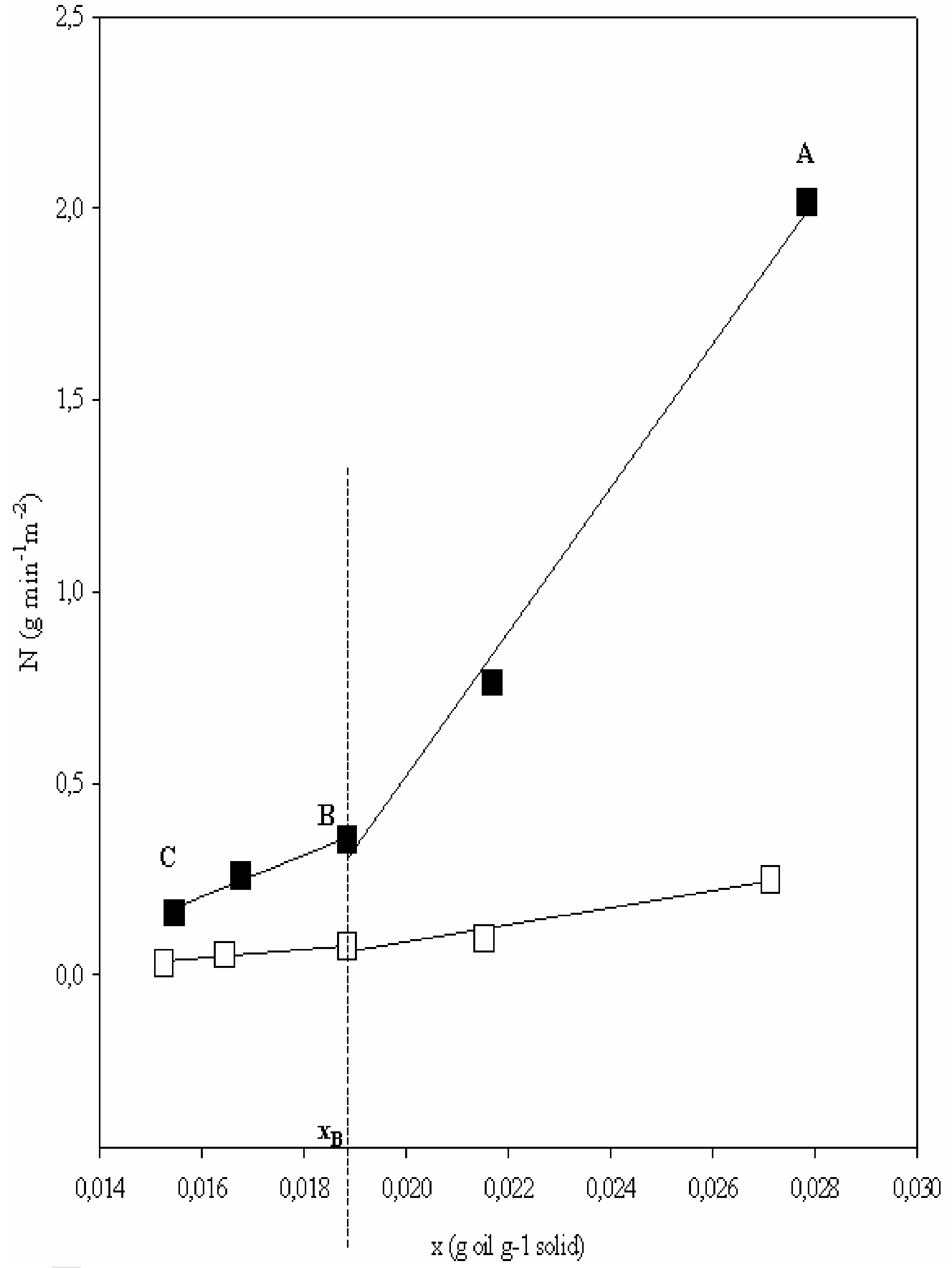
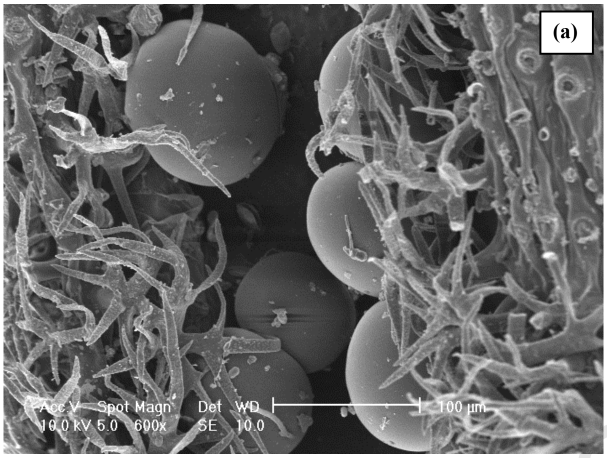
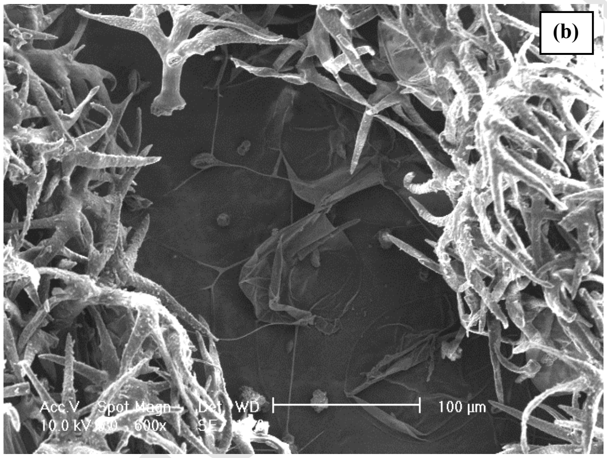

2,5
2,0
1,5
1,0
0,5
0,0
N (g min^{-1}m^{-2})
A
B
C
x_B
0,014
0,016
0,018
0,020
0,022
0,024
0,026
0,028
0,030
x (g oil g-1 solid)


(a)
Acc.V Spot Magn Det WD
10.0 kV 5.0 600x SE 10.0
100 µm


(b)
Acc.V Spot Magn Det WD
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100 µm

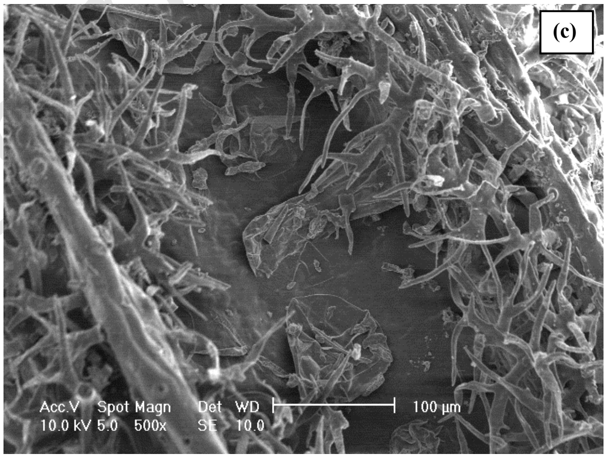
(c)
Acc.V Spot Magn Det WD
10.0 kV 5.0 500x SE 10.0
100 µm

Table.1 MSD experimental conditions for lavender extraction.

Parameters	G_v (g min^{-1})	P_w (W)
	3	50
	6	100
	25	200
	50	300
	60	400

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Table.2 Chemical composition of lavender essentil

n°	Compound	I^a	MSD (%)				SD (%)		
	Extraction time (min)		**1**	**2**	**3**	**5**	**10**	**15**	**20**
	Monoterpenes								
1	α- Thujene	937	0.154	0.175	0.094	0.049	0.059	0.158	0.0128
2	α-Pinene	937	0.51	0.6	0.32	0.161	0.88	0.025	0.083
3	Camphene	955	0.4	0.45	0.24	0.129	0.049	0.107	0.04
4	Sabinene	976	0.167	0.15	0.086	0.053	0.055	0.13	0.032
5	P -Pinene	971	1.1	0.87	0.53	0.467	0.24	0.35	0.138
6	β-Myrcene	990	0.69	0.446	0.347	0.336	0.74	0.566	0.02
7	α- Phellandrene	1010	0.069	0.057	0.034	0.029	0.04	0.137	0.13
8	p-Cymene	1030	0.12	0.091	0.058	0.04	0.04	0.067	0.048
9	limonene	1034	1.467	13.71	14.40	0.43	0.34	0.249	1.2
10	Eucalyptol	1038	11.659	7.83	4.9	4.69	3.62	4.17	2
11	cis-β- Ocimene	1049	0.266	0.149	0.146	0.169	0.43	0.226	0.125
12	γ-Terpinene	1063	0.236	0.179	0.116	0.098	0.078	0.114	0.068
	Oxygenated monoterpenes								
13	Terpinolene	1091	0.46	0.31	0.2	0.165	0.09	0.08	0.11
14	Linalool oxide	1093	0.668	1.044	0.47	0.46	0.6	0.67	0.08
15	Linalool	1104	35.51	30.47	27.1	19.09	44.86	32.73	28.88
16	Camphor	1160	8.923	9.255	8.152	8	6.25	7.44	2.5
17	Lavendulol	1170	0.432	0.747	1.119	1.07	0.317	1.08	0.339
18	Borneol	1184	9.411	5.438	6.225	6.4	4.3	4.58	2.12
19	Terpinen-4-ol	1191	5.795	7.245	7.47	7.24	6.5	7.29	2.71
20	α-Terpineol	1204	0.217	0.37	0.499	0.55	1.53	4.94	0.45
21	Geranyl acetate	1287	0.191	0.398	0.566	2.86	2.61	1.14	4.25
	Sesquiterpenes								
22	Caryophyllene	1435	0.664	1.31	1.84	1.6	1.17	0.47	2.29
23	(Z)-β-Farnesene	1457	0.644	1.59	2.65	2.38	1.84	0.72	3.63
23	Germacrene D	1497	0.26	0.679	1.37	1.23	0.88	0.2	1.7
25	Linalyl isovalerate	1505	0.106	0.287	0.673	0.67	0.71	0.40	1.18
26	α-Farnesene	1509	0.027	0.083	0.15	0.14	0.129	0.398	0.287
27	γ-Cadinene	1528	0.05	0.13	0.29	0.26	0.29	0.22	0.45
	Oxygenated sesquiterpenes								
28	P-Sesquiphellandrene	1534	0.33	0.119	0.27	0.26	0.25	0.081	0.42
29	Caryophyllene oxide	1601	0.096	0.119	0.432	0.57	0.699	0.52	1.44
30	Ť-Cadinol	1659	0.311	0.136	0.568	0.81	1.848	1.55	2.49
31	α-Bisabolo	1700	1.485	0.522	2.31	3.33	7.96	6.5	12
	Other oxygenated compounds								
32	3-Octanone	987	0.45	0.32	0.2	0.185	0.15	0.152	0.064
33	Terpenol(154)	1076	1.65	1.229	2.02	1.06	0.76	1.27	0.29
34	Propanoic acid, 2-methyl, hexyl ester	1149	0.435	0.4	0.323	0.27	0.178	0.05	0.22
35	Butanoic acid, hexyl ester	1193	1.19	1.36	1.21	1.141	3.566	4.024	1.109
36	Butanoic acid, 2-methyl-, hexyl ester	1283	0.255	0.295	0.318	0.249	0.2	0.328	0.33
37	Butanoic acid, 3-methyl-, hexyl ester		0.28	0.349	0.377	0.283	0.189	0.075	0.36
38	1,6-Octadien-3-ol, 3,7-dimethyl, acetate	1252	17.73	19.61	21.36	20.8	16.64	3.54	33.48
39	Cyclobutanecarboxylic acid, hexyl ester??	1334	0.191	0.398	0.566	0.48	0.35	0.3	0.7
40	Unknown diol	1337	0.037	0.131	0.355	0.58	0.46	0.52	0.097
	Extraction time (min)		1	2	3	5	10	15	20
	Yield (%)				4.42				4.42

(a) I: Re lative retention indices relatives to C_5-C_{28} *n*-alkanes calculated on non-polar HP5MS™ capillary coolumn.

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Table 3. Mass transfer results.

Parameters	MSD	SD
K (g m^{-2} min^{-1})	32.230	6.375
α (Bar^{-1})	0.38749715	0.18657825

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Table 4. Yields, extraction time and energy consumption of lavender flowers from extracted by MSD and SD.

	MSD	**SD**
Extraction time (min)	3	20
Yield (%)	4.42	4.43
Environmental impact (Kg CO_2 rejected. kg $^{-1}$oil)	69.3	338
Energy consumption (KWh. kg $^{-1}$oil)	86.42	423.83
Floral waters rejected (liter. kg $^{-1}$oil)	62.2	413

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