

Pilot Steam Distillation of Rosemary (*Rosmarinus officinalis* L.) from Portugal

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Abstract. While considering the project and the economic evaluation of an industrial steam distillation unit for essential oils there is a need to take into consideration that full extraction will need a lot of steam and that the length of the operation will determine the processing capacity of the unit. One must also quantify and dimension the equipment according to seasonal variations and associated processing peaks. Pilot tests are useful in obtaining data to cope with this problems and dimensioning industrial stills. Portuguese *Rosmarinus officinalis* L. is evaluated as an example.

Key words: essential oils; scale-up

Destilação por Vapor de Água de Alecrim (*Rosmarinus officinalis* L.) de Portugal

Sumário. Na elaboração do projecto e avaliação económica duma unidade industrial de destilação por vapor de água de óleos essenciais é necessário ter em consideração que a extracção total dos óleos é exigente no consumo de vapor e que a duração da operação determinará a capacidade de produção do destilador. É também necessário quantificar e dimensionar o equipamento de acordo com as variações sazonais de recolha da planta. Os testes piloto são importantes na obtenção de dados para a resolução destes problemas e dimensionamento de destilarias industriais. O alecrim (*Rosmarinus officinalis* L.) de origem portuguesa foi avaliado como exemplo.

Palavras-chave: óleos essenciais; aumento de escala

Distillation à la Vapeur d'Eau du Romarin (*Rosmarinus officinalis* L.) de Portugal

Résumé. Lors de l'élaboration du projet et de l'évaluation économique d'une unité de distillation d'huiles essentielles il faut prendre en considération que l'extraction exhaustive de

l'huile par distillation à la vapeur est exigeante en consommation de vapeur et que la durée de l'opération détermine la capacité productive du distillateur. Celle-ci sera aussi importante pour déterminer la dimension des équipements et des installations, en accord avec la saisonnalité de la récolte de la plante. Des essais-pilotes peuvent être utiles pour résoudre ces problèmes de dimensionnement des distillateurs. Le romarin d'origine portugaise (*Rosmarinus officinalis* L.) a été évalué à titre d'exemple.

Mots clés: huiles essentielles; augmentation d'échelle

Introduction

Batch distillation using steam passing through the vegetal charge in a still is the classical process for obtaining essential oils from medicinal and aromatic plants in industry. The steam mode is used in laboratory as well (WHISH, 1995; BOUTEKEDJIRET *et al.*, 1997; BOUTEKEDJIRET *et al.*, 2003) although the distillation mode where the plant is immersed in boiling water is more common (water distillation). Many alternative principles have been tried in innovative processes and a few new ones have been put in industrial or semi-industrial practice, supercritical fluid extraction being the most relevant. Anyway economics keep on putting a brake on many applications. It is mainly in the extractive methods for analytical purposes that proposals for more efficient and less time consuming methods seem more rewarding.

Being a safe and simple process that under most circumstances is compatible with the environment, steam distillation is the ideal process to use in areas where education levels are low. On the other hand these characteristics may explain why it took some time for this operation to deserve careful study. Everybody is able to put it in practice without giving it much thought especially when low capacities are concerned. Things change in the case of high capacities and when the efficiency of the processing

operations is important. It was not many years ago that DENNY (1988, 1991) proposed an elaborated and systematic approach to the essential oil distillation as an engineering operation, namely a methodology for scale-up from pilot-distillation tests.

Comparing the steam distillation practice in laboratory and industry some important differences should be put in perspective. Laboratory distillation aims to be exhaustive according to the ability of the used method, originating reproducible results while preserving the original components – KOEDAM (1982) emphasized the contribution of the operation to chemical changes in the original composition. On the other hand industrial operation doesn't have to be exhaustive according to the acceptable quality of the oil for the target applications. In the laboratory as a research tool steam distillation is often performed upon selected parts of the plant while in a industrial processing plants in general have to be accepted as a whole and as they were collected. In industry the economy of the process must be accounted for – time and spent energy being the most important parameters.

Steam flow used for distillation is quantitatively very ineffective in the last period of distillation, when little oil is obtained, time consumed and energy keep spending as when the operation started. One has to decide when to stop,

meaning that with shorter extraction periods the oil composition will be representative but not exhaustive and that such decision together with the packing density of the plant and the yield of oil will be important to establishing of equipment capacity and productivity. To estimate such values experiment data from pilot tests are a valuable contribution specially taking in consideration that the distillation capacity should match the peak availability of the plant according to its seasonal profile.

DENNY (1991) had reported as an example the use of his methodology to a *Labiatae*, a lavender. The same methodology has been used in the present case of portuguese rosemary (*Rosmarinus officinalis* L.) a perennial subshrub also belonging to the *Labiatae* family that grows wild in Portugal and in the coastal mediterranean regions. It can also grow as cultivars with an abundant flowering period after two to three years. Rosemary's oil finds application in perfumery, soaps, bath foams, shampoos, hair tonic lotions and as an odour mask (BAUER *et al.*, 1988). In food industries it is used in meat products and spices.

The methodology for pilot-tests

Distillation tests conducted according to the method proposed by DENNY (1991) are based upon the determination of two parameters (the "increment parameter" s and the "basic time parameter" t) after what dimensioning of a still suitable for the processing of the plant could be made.

Which is the meaning of these parameters? Assuming that oil glands after collapsing in contact with steam,

have the shape of a disk with area a and radius r and also considering that the use of a given steam flow oil will be extracted in time t proportional to radius $r = (a/\pi)^{1/2}$, the parameter t may be deduced as:

$$t = E^{-1} \left(\frac{a}{\pi} \right)^{1/2} \quad (1)$$

where E is a proportionality constant.

Then considering each charge height unit, area a will be incremented by δa with the upper layers of the charge receiving oil from the lowers. A charge with height H will have at its top oil disks with an area $a + H \cdot \delta a$. Therefore, extraction time would be:

$$T = E^{-1} \left(\frac{a + H \delta a}{\pi} \right)^{1/2} \quad (2)$$

Performing tests using two charges with similar packing densities, the increment parameter s (defined as $s = a/\delta a$) can be determined comparing extraction times when height changes from H to H' as:

$$\frac{T'}{T} = \left(\frac{s + H'}{s + H} \right)^{1/2} \quad (3)$$

A similar development using equations (1) and (2) and the definition of the increment parameter s , gives the basic time parameter t (the time necessary to reduce to nil de radius of each oil disk of area a to nil) as:

$$t = T \left(\frac{s}{s + H} \right)^{1/2} \quad (4)$$

Using the values of s and t , any extraction time as a function of the height of the charge can now be estimated:

$$T = t \left(1 + \frac{H}{s} \right)^{1/2} \quad (5)$$

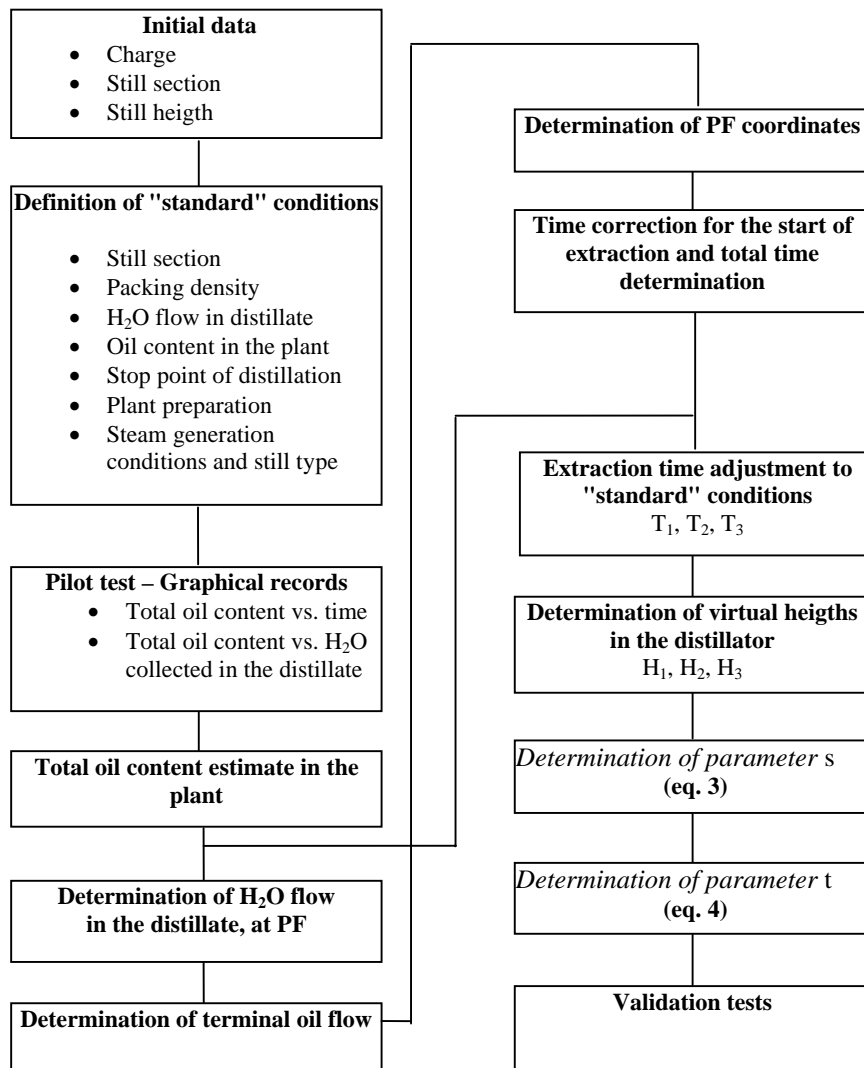
For a given plant, the parameter t should be adjusted to different pilot-test's conditions since it is inversely proportional to the steam velocity while

the parameter s varies inversely with the oil content of the plant.

For each plant with oils located on absorbing surfaces, several distillation tests using charges with different heights in the still are performed - generally three - one of them being used to

establish "standard" conditions. Reduction of each test to this standard condition will make the comparison between tests possible.

The methodology for pilot distillation tests with plants having superficial oils is presented in Figure 1.



PF – final point of distillation

Figure 1 - Methodology of pilot distillation tests

Experimental

Plant material and drying process

The plant was collected on two consecutive years, during July, near Évora, Portugal. These lots will be from now on referred as lot A and lot B. The plant was cut in a way simulating the use of mechanical harvesting equipment (leaves and caulis together). The first lot was dried under roof at room temperature for 22 days and then distilled while the second lot was distilled the day after being collected.

Pilot-still operating conditions and facilities

Tests were carried in an inox still built with two cylindrical bodies with 0.24 m² of cross-section the lower one with a perforated support for the plant above the steam injection. The condenser had 17 tubes 1 m long with internal diameter of 17 mm discharging the condensate in a separator - initially filled with water - where oil accumulated in a graduated cylindrical tube.

Steam was generated in a boiler (VAPORAX 150) with 150 L/h capacity at 10 bar. Pressure was reduced to 3 bar on its way down to the still. Pressure and temperature in the injection line were recorded. During the distillation of the lot A pressure readings were between 1.9 bar and 3.1 bar with mean value of 2.3 bar (gauge pressure). Temperature readings were between 128°C and 136°C. Water flowing out the separator was gathered in a recipient and accumulated value read at regular intervals.

Some modifications have been made for the distillations of lot B. The flow of steam was stabilized by a dampening

chamber before injection. The pressure inside the dampening chamber varied between 1.4 bar and 1.6 bar (gauge pressure) which was a narrower range than that obtained in the direct injection line. Steam temperature oscillated between 122°C and 130°C.

Oil was recovered in a graduated cylindrical tube and accumulated volume read at regular intervals. Since it was difficult to read simultaneously two oscillating levels for lot B the accumulation was recorded on video and measurements taken from the frozen image.

All distillations were made with a charge height of about 1/3, 1/2 and 2/3 of the total height of the still.

Results and discussion

Pilot-distillation tests

In Table 1 data on the distillations of lots A and B are shown. Curves that represent the accumulated oil evolution obtained during the tests all show the same sigmoidal profile characteristic of oil glands located mostly on the surface of the plant (see example in Figure 3).

Tests for checking the location of oil glands in the plant are indicated by DENNY (1991). In one of them whenever two identical charges are processed in the same still using two different flows of steam and if time of extraction varies inversely with flow of water in the distillation that may indicate positively that oil glands are located at the surface. Proportionality seems better the greater the charge tested. In the other test when representing the ratio volume of condensed steam/oil volume as a function of the volume of the total

condensed steam one should verify that all curves would develop upwards from a certain initial level with a more or less pronounced elongation according with total quantity of essential oil.

These tests seem useful for a first insight when no other knowledge about the plant physiology is at hand which is not the case with rosemary where the superficial location of the oil glands is well known. Anyway we checked them and no conclusion on the location of oil was possible on the basis of proportionality between steam flow and duration of distillation for lot B. A possible cause could be the small variation on steam flow from test to test (maximum ratio of

1.1:1) the changes in the charge being much more significant (29.9 kg to 11.55 kg maximum ratio of 2.6:1). For the second test as exemplified with lot B tests in Figure 2 the results agree with the behavior indicated for plant with oil superficially located. Curve for test designated as EMM032 behaves in a slightly different way from the others very likely due to the small height used causing a longer path for steam and oil after passing through the plant and before leaving the equipment. In this case condensation along the metallic walls of the still would slow down the oil progression giving higher water/oil ratios coming out of the condenser.

Table 1 - Distillation of rosemary lots. Experimental results (lot A and lot B)

Height (cm)	Plant weight (kg)	Packing (kg/m ³)	v steam (L/min. m ²)	V oil (mL)	m oil (g)	d oil (g/mL)	η (% m/m)	Tag	
46	10.70	96.9	3.09	65	54.4	0.84	0.51	Al ₁	LOT A
47	9.90	87.8	3.00	65	54.3	0.84	0.55	Al ₂	
47	9.90	87.8	2.80	74	62.1	0.84	0.63	Al ₃	
72	17.85	103.3	3.13	126	105.2	0.83	0.59	Al ₄	
70	14.50	86.3	2.99	104	88.3	0.85	0.61	Al ₅	
70	10.85	47.6	2.92	82	68.8	0.84	0.63	Al ₆	
72	17.55	101.6	2.89	132	110.8	0.84	0.62	Al ₇	
95	22.35	98.0	2.99	174	147.4	0.85	0.66	Al ₈	
96	22.95	99.6	3.72	192	162.3	0.85	0.71	Al ₉	
95	21.10	92.5	2.92	164	138.4	0.84	0.66	Al ₁₀	
95	26.45	115.1	3.09	173	148.7	0.86	0.56	EMM028	LOT B
95	29.90	130.1	3.14	181	155.4	0.86	0.52	EMM029	
71	22.35	130.1	3.07	154	132.5	0.86	0.59	EMM030	
71	20.40	118.8	3.16	119	101.4	0.85	0.50	EMM031	
47	11.55	101.6	3.19	61	52.5	0.86	0.46	EMM032	
42	8.60	84.6	3.45	30	25.9	0.86	0.30	EMM033	

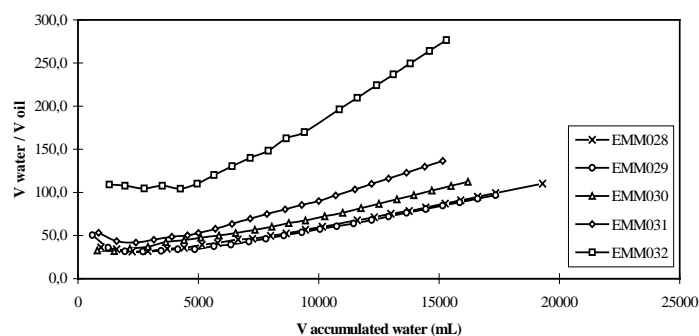


Figure 2 - Evolution of the water-oil ratio (v/v)

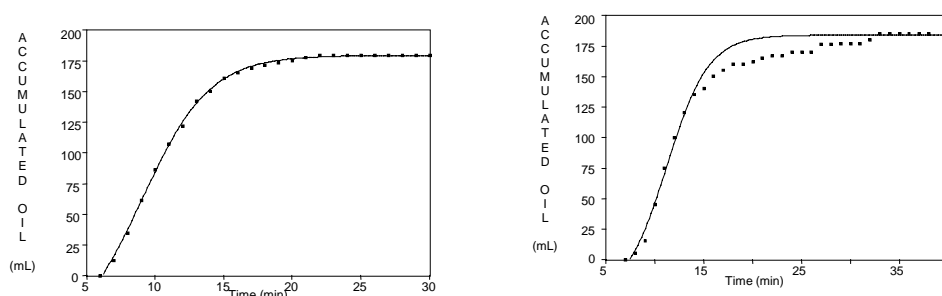


Figure 3 - The curve on the left corresponds to test EMM029 (lot B) and the curve on the right to test Al 9 (lot A). It's represented the deviations between estimated and observed distillation time

Extraction time estimation. Determination of model parameters

The main objective of the steam distillation tests consists in determining the values of the incremental parameter s and the basic time parameter t which should allow us given the steam flux to estimate the processing time of charges with a given height in a scale-up calculation.

For the determination of the characteristic parameters s and t , the beginning of the extraction period was taken as the moment the first drop of oil

coming out of the condenser was observed. The end of the extraction was set in two different ways. For tests in lot A, the end of the extraction was set when the distillation rate attained 3.6 mL/minute corresponding to the distillation of 95% of the total oil while for the tests made with lot B the end of the extraction was set at 98% of the total oil. The corresponding time was determined through distillation curves similar to those represented in Figure 3 fitting the curve to experimental values and its first derivative used to determine the end point.

In Table 2, calculated values of s and t obtained from the results of two tests with lot A (Al 5 corresponding to a still charge of $\frac{1}{2}$ of its height and Al 1 corresponding to $\frac{1}{3}$ rd of its height), the Al 5 test was taken as the standard. In the same Table, where these parameters have been used to estimate extraction times for the remaining tests, mean errors between estimated and measured times (corrected for standard conditions) are indicated. Some significant deviations between estimated and observed distillation time can be noticed specially for the higher charges. These were mainly originated by drop retention inside "florentino flask" which caused distortion in the curve representing the accumulated oil *versus* time (Figure 3, test Al 9), thus affecting the terminal rate and the corresponding time determination which value is used in the estimate calculations. The distortion is greater the greater the charges and the greater the oil volume retained inside the apparatus.

Other factor causing difficulties in the conduction of the tests were the measurement of the accumulated oil and the oscillation of pressure in the injection line, which made difficult to assign a

value to the steam rate. With improvements made in the equipment for processing lot B, steam pressure was easier to maintain and steam rates varied between 3.09 and 3.45 L/min/m².

Any reasonable conditions can be set as standard according to the used methodology. After making the proper corrections to the still's charge height and to the distillation time parameters, s and t can be determined. Anyone may quest which is the best combination of tests for the best fitting of experimental distillation times. Table 3 indicates mean errors in the estimation of distillation times using parameters s and t calculated according to standard and auxiliary tests used. The best pair is the one that uses test EMM028 as standard and EMM032 as auxiliary. All possible combinations in Equation (1) were tried excluding crossed combinations from a test with itself which are obviously meaningless, and the pairing of tests with similar heights which are inadequate to the use of Equation (1). Some pairings originate nonsense s parameters (negative). As we show, these pairings may be ruled out by previous inspection because they violate validity conditions for the relationship of results obtained.

Table 2 - Tests with lot A: parameters s and t determined and estimates of extraction times based upon them ("Standard" test: Al 5; Auxiliary test: Al 1)

Test	T extraction corrected (min)	H corrected (cm)	s	t	T extraction calculated (min)	Error (%)
Al 1	6.9	44.4			6.9	0.0
Al 2	6.7	43.8			6.8	1.3
Al 3	6.3	49.8			7.1	13.4
Al 4	8.7	84.8			8.7	-0.6
Al 5	8.0	67.5	23.3	4.0	8.0	0.0
Al 7	10.6	82.1			8.6	-19.2
Al 8	12.5	117.1			9.9	-20.8
Al 9	14.8	129.2			10.3	-30.4
Al 10	13.3	110.4			9.7	-27.2

Table 3 - Mean errors in the estimation of distillation time as a function of chosen standard and auxiliary tests (lot B). (H_t = total height of the still)

Standard test	Auxiliary test for s and t determination				
	EMM028 (2/3 H_t)	EMM029 (2/3 H_t)	EMM030 (1/2 H_t)	EMM031 (1/2 H_t)	EMM032 (1/3 H_t)
EMM028 (2/3 H_t)			12.99%		8.55%
EMM029 (2/3 H_t)				13.55%	8.62%
EMM030 (1/2 H_t)					9.78%
EMM031 (1/2 H_t)					18.29%

As a matter of fact, s may be calculated from equation (3) as:

$$s = \frac{H_2 - \left(\frac{T_2}{T_1}\right)^2 \cdot H_1}{\left(\frac{T_2}{T_1}\right)^2 - 1} \quad (5)$$

Then, s will be positive if the following conditions are satisfied:

$$T_2 > T_1 \quad \Rightarrow \quad \frac{H_2}{H_1} > \left(\frac{T_2}{T_1}\right)^2 \quad (6)$$

$$T_2 < T_1 \quad \Rightarrow \quad \frac{H_2}{H_1} < \left(\frac{T_2}{T_1}\right)^2 \quad (7)$$

None of the pairing conducting to negative s parameters satisfies those conditions and they were not considered.

In Table 3 mean errors obtained for estimated extraction times are indicated for each different pairs of tests used to calculate the parameters s and t - one used as standard and the other as auxiliary. Each pair s and t will define a function allowing the estimation of distillation time as a function of charges height. For each pairing, extraction times are estimated for all the remaining tests with the exception of those used as standard and auxiliary.

In Figure 4, curves corresponding to functions obtained for parameters resulting from pairing tests as indicated are shown. The curve for the mean estimation of all possible pairings is also shown. These curves indicate that 30 minutes should be enough with some confidence margin to extract charges with height up to 2 m as long as steam flux is adequate.

RAO (1999) reported a field distillation using a 1000 kg capacity distillation vessel using steam at about 2 bar. It took 120 minutes to what was considered a full extraction and about 60 minutes to obtain 90% of the total oil extracted. No data is given about steam flux or about the height of the charge.

Oil accumulation model and distillation rate

Taking the equation found to fit the oil accumulation data during test EMM029, its derivative representing the distillation rate was analytically calculated. Both curves - oil accumulation and distillation rate - are represented in Figure 5. A maximum on the distillation rate was found at 9.1 minutes after the steam injection began. Removing the lag

time taken by the condensed steam and oil to flow out of the condenser the maximum corresponds to about 3 minutes after distillation started. This curve corresponds well in the maximum and in the shape to the curve obtained by BOUTKEDJIRET *et al.* (2003) in a laboratory steam distillation of rosemary taking in consideration that heat losses

through walls are much more important in small apparatus than in bigger ones, making the oil progression to the condenser slower and that steam conditions and rate are milder in laboratory. This matching indicates that the oil release in the plant follows a similar pattern in time in both systems independently of the scale.

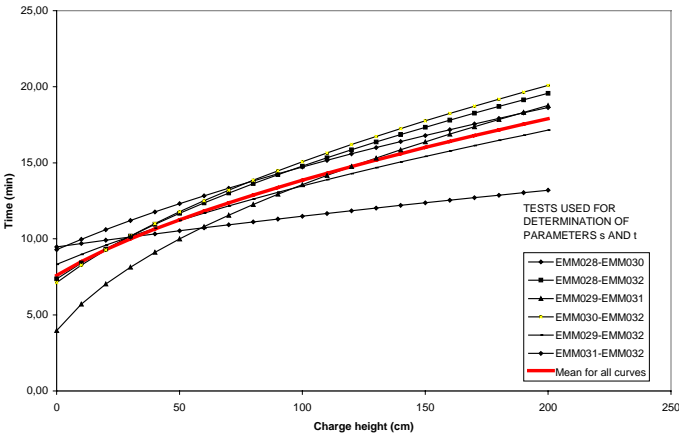


Figure 4 - Distillation time estimation vs. charge height as a function of (s, t) parameters

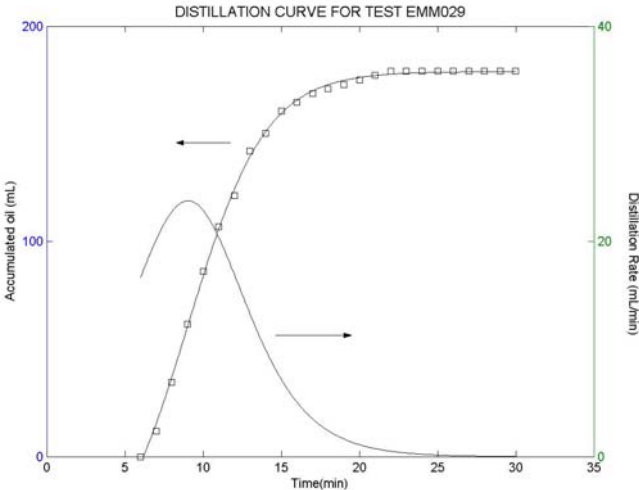


Figure 5 - Oil accumulation and distillation rate with test EMM029. Arrows indicate the ordinate axes

Chemical composition of essential oils

Samples of the essential oils were analyzed by gas-liquid chromatography in a Carlo Erba 6000 Vega, Series 2 apparatus, with a FID detector and using a DB-1 column (30 m x 0,32 mm x 0.25 μ m) from JW and a Carbowax 20 M column (30 m x 0.25 mm x 0.25 μ m) from Restek.

When using the DB-1 column, the injector temperature was 230°C and the detector 240°C. The program temperature started at 45°C ramping to 175°C at a rate of 3°C/min. Another ramp followed from 175°C to 240°C at a rate of 15°C/min. Helium was the carrier gas (1 mL/min). With the Carbowax column the injector temperature was 200°C and the detector temperature 240°C. The temperature program used a ramp from 70°C to 200°C at a rate of 2°C/min. The carrier gas was hydrogen (1 mL/min).

In Table 4, composition analysis results for lot A on the DB-1 column are shown while the results obtained on the Carbowax column are in Table 5. On both tables, the column at right shows the results for a sample obtained in a Clevenger using the same plant material. Compounds were identified by comparison with standard retention indexes from bibliography and with standard samples for myrcene, α -pinene, α -humulene, β -pinene, 1,8-cineol, linalol, camphor, borneol and limonene.

The analysis of the samples of the essential oil extracted during the tests showed significant contents on two monoterpene hydrocarbons α -pinene and myrcene. A high content on myrcene in oil from portuguese origin had been indicated by CARDOSO DO VALE *et al.*

(1980), and later confirmed by PROENÇA DA CUNHA *et al.* (1986), LAWRENCE *et al.* (1993) and SERRANO *et al.* (2002). Camphor and 1,8-cineole were the two more important oxygenated monoterpenes. Compositions obtained for all samples demonstrated similar pattern despite of the different heights tested (Figure 6). Lot A has a mean content of 35.5% for myrcene and of 11.7% for α -pinene while for referred oxygenated compounds mean contents are 13.1% for 1,8 cineole and 9.5% for camphor. For lot B, contents are 34.4%, 12.4%, 12.9% and 10.4% respectively for myrcene, α -pinene, 1,8-cineole and camphor, respectively.

Comparing the composition of the essential oils obtained in this study with those obtained in Clevenger, higher contents in myrcene and β -caryophyllene were observed while contents in camphor and 1,8-cineole decreased (Figure 7), which should be caused by a greater relative mass of water contacting the oil.

Conclusions

The useful information provided by pilot scale experiments is not confined to yields and compositions of essential oils. They allow estimates on distillation times and these are very important when specifying plant and utility capacities to cope with the flux of vegetal material at harvest. In industry, while keeping the oil within its specifications, extraction times should be close to the minimum necessary for the previous reason, but mainly to economize energy and improve the productivity of the equipment.

Table 4 – Chemical composition of essential oils obtained from rosemary's pilot distillations with lot A (DB-1 column)

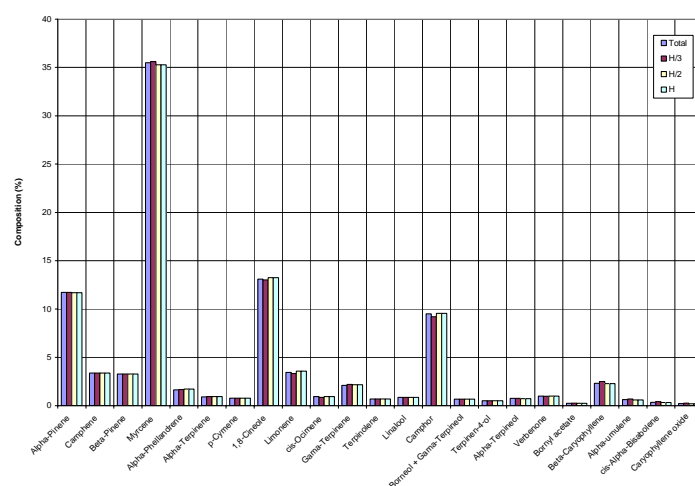
N.	Componente	RI DB-1	RI CW 20M	Al 1	Al 2	Al 3	Al 4	Al 5	Al 6	Al 7	Al 8	Al 9	Al 10	Al Cl
1	α -Pinene	933	1027	12.1	11.6	11.4	12.6	11.9	11.1	11.3	12.2	11.8	11.2	11.2
2	Camphene	943	1072	3.4	3.4	3.4	3.5	3.4	3.3	3.3	3.3	3.3	3.6	3.4
3	β -Pinene	970	1113	3.5	3.2	3.1	3.5	3.2	3.1	3.3	3.4	3.2	3.2	2.9
4	Myrcene	990	1170	36.2	35.9	34.8	35.5	36.8	35.0	33.9	34.9	36.4	35.8	31.5
5	α -Phellandrene	996	1170	1.6	1.6	1.8	1.4	1.4	1.8	2.2	1.5	1.4	1.6	1.4
6	α -Terpinene	1008	1182	0.9	1.0	1.0	0.8	0.9	1.0	1.9	0.8	0.8	0.9	0.9
7	p-Cymene	1011	1268	0.7	0.8	0.9	0.7	0.7	0.8	0.9	0.8	0.9	0.8	1.4
8	1,8-Cineole	1021	1213	13.2	12.7	13.1	13.3	13.3	13.3	13.2	12.8	12.9	13.1	14.9
9	Limonene	1023	1201	3.3	3.3	3.4	4.0	3.4	3.3	3.6	3.2	3.4	3.5	3.2
10	cis-Ocimene	1032	1234	0.9	0.9	0.8	0.9	1.1	0.8	0.9	1.0	0.9	1.2	0.7
11	γ -Terpinene	1052	1245	2.2	2.2	2.2	2.0	1.9	2.3	2.5	2.0	1.9	1.9	1.8
12	Terpinolene	1079	1278	0.7	0.7	0.7	0.7	0.8	0.7	0.7	0.7	0.7	0.7	0.6
13	Linalool	1088	1549	0.9	0.8	0.9	0.9	0.8	0.9	0.9	0.9	0.9	0.8	1.2
14	Camphor	1118	1497	8.7	9.3	9.7	8.8	9.2	10.0	10.1	8.9	9.2	10.9	14.4
15	Borneol + δ -Terpineol	1146	1698 +1664	0.7	0.7	0.7	0.7	0.7	0.7	0.6	0.7	0.6	0.8	1.0
16	Terpinen-4-ol	1160	1590	0.5	0.5	0.5	0.5	0.5	0.6	0.6	0.5	0.5	0.5	1.0
17	α -Terpineol	1171	1676	0.7	0.8	0.8	0.7	0.7	0.8	0.8	0.8	0.7	0.8	1.4
18	Verbenone	1176	1730	1.0	0.9	1.0	1.1	0.9	1.0	0.9	1.2	1.0	1.0	2.6
19	Bornyl acetate	1265	1565	0.3	0.3	0.3	0.2	0.2	0.3	0.2	0.2	0.2	0.2	0.2
20	β -Caryophyllene	1420	1593	2.3	2.6	2.6	1.6	2.2	2.8	2.5	2.2	2.0	2.2	0.5
21	α -Humulene	1439	1642	0.7	0.7	0.7	0.4	0.6	0.8	0.6	0.6	0.5	0.6	0.1
22	cis- α -Bisabolene	1496	1736	0.4	0.4	0.4	0.2	0.3	0.4	0.4	0.3	0.3	0.3	0.1
23	Caryophyllene oxide	1555	1943	0.3	0.3	0.3	0.1	0.2	0.3	0.2	0.2	0.2	0.2	0.2
24	Non-identified			5.1	5.5	5.7	5.9	5.0	5.3	5.2	7.2	6.3	4.4	3.6

Bold numbers - percentages > 5%

Table 5 – Chemical composition of essential oils obtained from rosemary's pilot distillations with lot B (DB-1 column).

No.	Component	RI DB-1	RI CW 20M	EMM0 28	EMM0 29	EMM0 30	EMM0 31	EMM0 32	EMM0 33
1	α -Pinene	933	1027	12.5	12.8	12.8	12.3	11.5	12.8
2	Camphene	943	1072	3.5	3.5	3.5	3.5	2.8	2.8
3	β -Pinene	970	1113	3.2	2.8	2.9	2.5	2.7	2.8
4	Myrcene	990	1170	33.2	35.2	35.0	35.0	33.0	34.8
5	α -Phellandrene	996	1170	1.8	1.7	1.4	1.3	1.8	1.7
6	α -Terpinene	1008	1182	0.8	1.0	0.7	0.7	0.9	0.9
7	p-Cymene	1011	1268	1.3	1.6	1.4	2.4	1.5	1.3
8	1,8-Cineole	1021	1213	12.6	13.1	12.5	12.9	12.9	13.3
9	Limonene	1023	1201	4.5	3.6	3.3	3.6	5.1	5.3
10	cis-Ocimene	1032	1234	1.3	1.1	1.0	0.8	1.9	1.6
11	γ -Terpinene	1052	1245	1.7	2.1	1.7	1.4	2.1	2.0
12	Terpinolene	1079	1278	0.6	0.6	0.5	0.5	0.7	0.7
13	Linalool	1088	1549	1.0	0.9	1.0	0.9	1.0	0.9
14	Camphor	1118	1497	11.1	9.9	11.3	11.5	10.1	8.6
15	Borneol + δ -Terpineol	1146	1698 + 1664	0.8	0.7	0.8	0.8	0.9	0.7
16	Terpinen-4-ol	1160	1590	0.6	0.6	0.6	0.6	0.7	0.6
17	α -Terpineol	1171	1676	0.8	0.8	0.9	0.8	0.9	0.8
18	Verbenone	1176	1730	1.1	1.1	1.1	1.4	1.1	0.8
19	Bornyl acetate	1265	1565	0.3	0.3	0.3	0.4	0.3	0.3
20	β -Caryophyllene	1420	1593	1.7	1.7	1.9	1.5	2.3	2.4
21	α -Humulene	1439	1642	0.4	0.4	0.5	0.4	0.6	0.6
22	cis- α -Bisabolene	1496	1736	0.2	0.2	0.3	0.3	0.4	0.4
23	Caryophyllene oxide	1555	1943	0.1	0.1	0.2	0.2	0.3	0.2
	Non-identified			4.9	4.1	4.6	4.3	4.6	3.5

Bold numbers - percentages > 5%

**Figure 6** - Mean total composition and mean composition for runs using full height of the still (H), half-height(H/2) and one third (H/3)

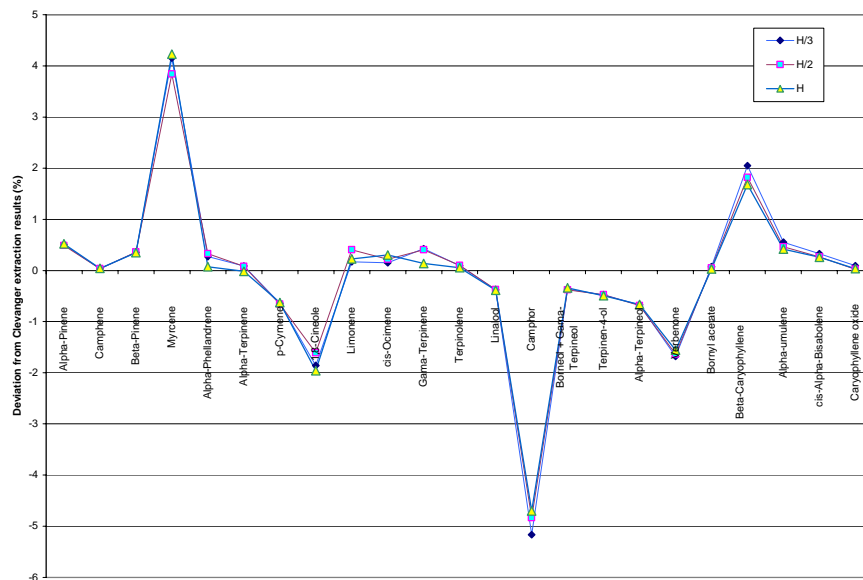


Figure 7 - Deviations from mean compositions for tests at different heights in the still - full height (H), half-height(H/2) and one third (H/3) - towards composition of the oil obtained in the Clevenger

Comparison between oil compositions extracted in laboratory and during pilot tests show that hydrophilic compounds, namely oxygenated compounds that are important for the quality of the oil, may have a significant decrease in the scale-up of the extraction. Prolonged contact between the oil and the passing condensed steam should be avoided. Oil compositions obtained in pilot tests showed consistency despite different heights of the charges. This consistency was favored by an adequate steam flow to avoid reflux.

For reliable results in the extraction times, estimation test conditions must be carefully defined. Results are sensitive to the use of small charges, being advisable not to rely on them in the determination of dimensioning parameters. A simple

check of experimental data, based on the preservation of the physical meaning of the underlying model assumed in the distillation tests, is advisable. Data not compatible should be discarded.

The comparison between data obtained in laboratory and in the pilot still for steam distillation of rosemary indicates the possibility of using the laboratory data on the rate of oil accumulation if properly adjusted to predict the still behavior as long as lags imposed by the dimensions of the equipment and the steam velocity differences are taken in consideration.

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