

Distillation Parameters for Pilot Plant Production of *Laurus nobilis* Essential oil

Temel Özek*

Department of Pharmacognosy, Faculty of Pharmacy, Anadolu University, 26470 Eskişehir,
Türkiye

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Abstract: Essential oils have increasing importance in flavour and fragrance industries. They are obtained by distillation techniques. In order to produce an oil with market potential its optimum production parameters have to be well known prior to its commercial production. Determination of the steam distillation parameters of commercially available Laurel leaves oil in pilot plant scale is described. The effect of steam rate and processing time play a major role in distillation of essential oils. Distillation speed was high in the beginning of the process, then gradually reduced as the distillation proceeded. The main component of the oil of Laurel leaf oil was 1,8-cineole accumulating significantly in the early fractions.

Keywords: Essential oil; processing; distillation parameter; Lauraceae; *Laurus nobilis*; laurel.

1. Introduction

Essential oils obtained by distillation from aromatic plant materials are complex mixtures of compounds of widely differing composition and boiling points and form very important constituents of cosmetics, perfumes, spices which contribute to fragrance, flavour and preservation of foods. These oils composed mainly of terpene hydrocarbons, sesquiterpenes and oxygenated components are enclosed in “oil glands” or “glandular hairs” in various parts of aromatic plants.

There are several techniques for isolating volatiles from plant materials. The basic methods are: water distillation, steam distillation, water and steam distillation, hydrodiffusion, etc. These techniques are described by Denny and Lawrence in detail [1, 2]. Commercial essential oils are obtained mainly by steam distillation. The process of steam distillation is the most widely used and accepted process for the production of essential oils on a large scale. However, for some commercial oils such as rose oil water distillation is employed. In cottage industries mainly in developing countries, water and steam distillation is practiced in which steam is generated by solid fuel at the bottom of the distillation still [1, 2].

The methods and equipments employed for the recovery of essential oils in developing countries are often obsolete, producing in poor and inconsistent quality of essential oils resulting in

*Corresponding author: E-mail address: tozek@anadolu.edu.tr Tel.: +90-222-3350580 / 3719; fax:+90-222-3306809

poor returns. The most commonly practiced and simplest of all the operations followed in the recovery of essential oils from the plant materials is steam distillation.

Steam distillation briefly is a unit operation in which live steam is passed through a well packed bed of plant material in a cylindrical still. The resultant vapours containing volatile essential oils and steam are condensed in a heat exchanger and the condensate forming two immiscible layers are separated into essential oil and water in a separator. The latter is called aromatic water (or *hydrosol*) of that plant and is usually discarded if it has no use or commercial value.

Although distillation is a well known and simple process, data on optimum conditions such as time of distillation, steam rate, etc. need to be worked out on individual raw materials, since every plant material is unique by itself and the conditions cannot be generalized.

In this study, the influence of distillation parameters such as time and steam rate on *Laurus nobilis* oil recovery on pilot plant scale were investigated.

2. Materials and Methods

2.1. Plant Material

Laurel (*Laurus nobilis* L.) grows along the entire coast line of Turkey at an altitude of 1200 m [3,4].

Laurel leaves are collected for export and the production of essential oil. The main collection areas are Aegean and the eastern Mediterranean regions, while there is limited production in the Black Sea region. It is also cultivated in the Hatay and İçel provinces mainly for its oil-bearing berries [4].

First grade standardized leaves are exported in special boxes after cleaning and sorting for use as a spice. The essential oil of Laurel leaves generally produced from the second grade leaves. In 2002, Turkey exported 4,869 tons of Laurel leaves for a return of USD 7.7 million with USD 1.58 unit export price [4].

Previous studies have indicated that the main component was 1,8-cineole which varied in the range from 23-63 % according to the different extraction techniques [5-37].

2.2. Steam Distillation of *Laurus nobilis*

Bench scale data on essential oil yield and moisture content of the plant material were completed before scaling up to pilot plant. In the laboratory, a Clevenger type apparatus was used to obtain essential oil for 3 hours by hydro-distillation. For moisture content determination, a glass apparatus was used and the method described in European Pharmacopoeia (2005) was followed [38]. Approximately 100 g plant material for hydro-distillation and 10 g for volumetric moisture content determination were taken from each batch.

Laurel leaves were collected from south-western part of Turkey. It contained 1.8-3.9 % total essential oil. Oil yields were calculated on moisture free basis. Overall relative oil recovery calculations were then based on the ratio of the oil volume obtained on pilot plant scale over that obtained on bench scale.

Pilot plant scale steam distillation experiments were conducted in a 500 Liter capacity stainless steel distillation unit. Steam was generated by a steam boiler fired by natural gas. Four batches were worked up in the pilot plant. The results are summarized in Table 1.

All the calculations were done by the following equations:

$$m = m_w (1-w) \quad (1)$$

$$Y_L = \frac{V_{it}}{m} \quad (2)$$

$$Y_P = \frac{V_T}{m} \quad (3)$$

$$Y_o = \frac{Y_P}{Y_L} \quad (4)$$

$$Y_R = \frac{Y_{Pt}}{Y_L} \quad (5)$$

$$V_R = \frac{V_t}{V_T} \quad (6)$$

$$R = \frac{V_t}{mt} \quad (7)$$

$$V_{OR} = \frac{V_D}{V_T} Y_R \quad (8)$$

For laboratory scale oil yield (Y_L) calculation, V_T assumed equal to V_{it} which is recovered at infinite time because of the negligible amount of oil observed at the end of the process.

Table 1. Experimental data of *Laurus nobilis*

| | Experiments | | | |
|---|-------------|----------|-----------|----------|
| | Batch I | Batch II | Batch III | Batch IV |
| Weight of batch; m_w (kg) | 24.5 | 34 | 38 | 38 |
| Moisture content; w (%) | 8 | 8 | 8 | 8 |
| Steam rate; R_{stm} [(kg steam) (kg material) ⁻¹ (h) ⁻¹] | 0.9-1.1 | 1.4-1.5 | 0.78-1.1 | 0.5-1.2 |
| Oil yield (Y_P) in Pilot scale; (L oil / kg material) (%) | 2.51 | 1.04 | 1.86 | 3.63 |
| Oil yield (Y_L) in Lab. scale; (L oil / kg material) (%) | 2.85 | 1.80 | 2.10 | 3.86 |
| Overall oil yield ratio (Y_o) (%) | 88.10 | 57.78 | 88.57 | 94.04 |

Figure 1 shows overall relative oil yield (%) while Figure 2 shows the steam rate effect on overall oil yield (%) in individual batches.

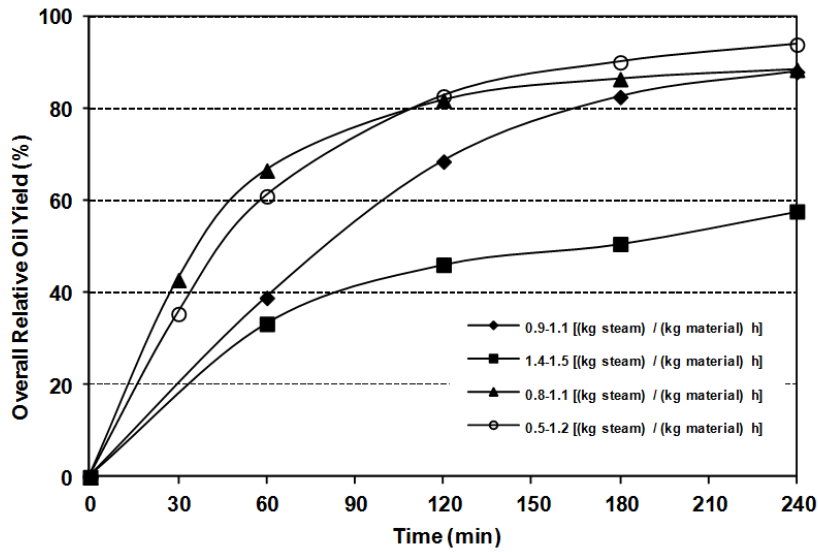


Figure 1. Overall relative oil yields of *Laurus nobilis*

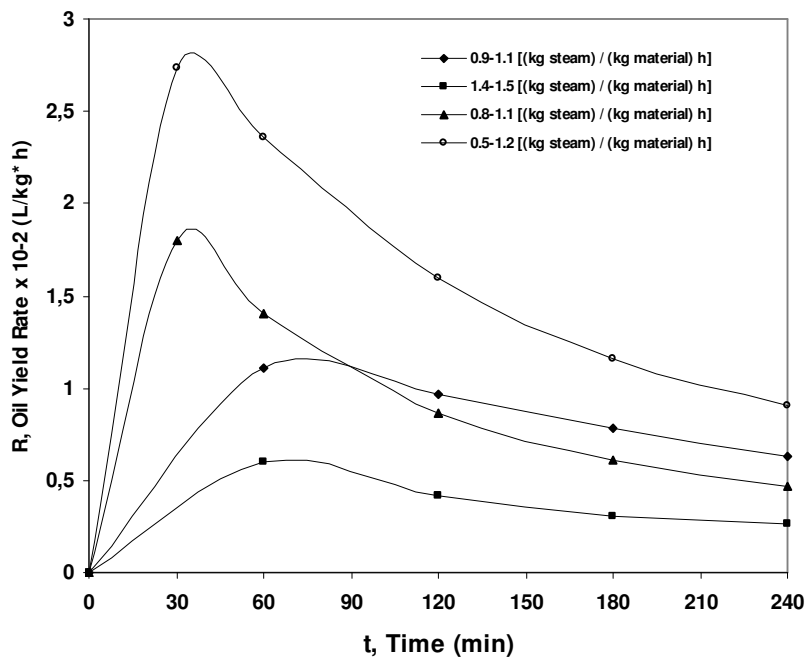


Figure 2. Steam rate effect on the yields for *Laurus nobilis*

Figure 3 shows oil percentages of each batch taken in differential time.

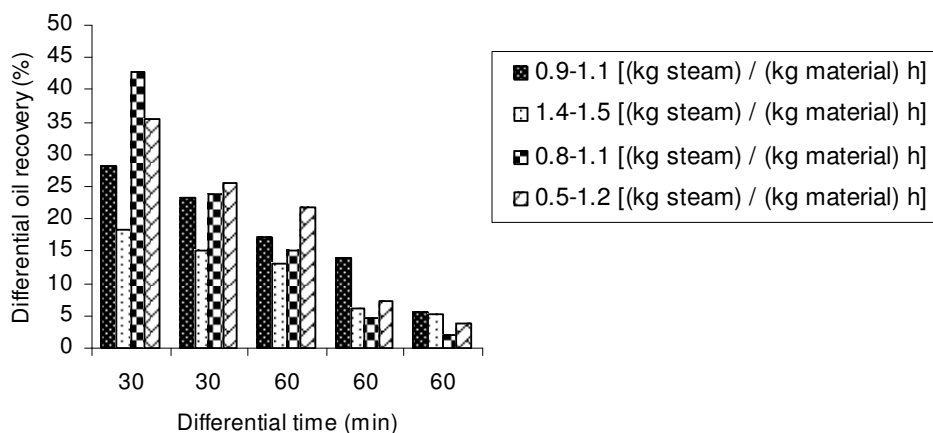


Figure 3. Oil distributions in differential time periods for *Laurus nobilis*

2.3. GC and GC/MS Conditions

The oil samples which were taken from each batch in fractions were analyzed by GC and GC/MS techniques. The gas chromatographic analysis was performed using a Shimadzu GC-9A with CR4A integrator with nitrogen as carrier gas. Oven temperature was held at 70°C for 10 min. and programmed to 180°C at a rate of 2°C/min, then held at 180°C for 30 min. Injector and detector (FID) temperatures were kept at 250°C. Thermon 600T fused silica capillary column (50 m x 0.25 mm id. with 0.25 µm film thickness) was used. Gas chromatography/mass spectrometry was carried out with Shimadzu GC/MS QP2000A system. The same column employed in GC analysis with the same conditions was used with helium as carrier gas at 0.8 atm. Identification of individual components was achieved using LSS-30 library search software from the “NBS/NIH/EPA Library”, “BASER Library of Essential Oil Constituents”, The Wiley/NBS Registry of Mass Spectral Data”

Composition of *Laurus nobilis* essential oil worked in this study is given in Table 2.

3. Results and Discussion

The study indicated that the steam rate is a very important parameter. Figure 1 shows the overall recovery of oil against the time of distillation for different steam rates applied. It is clear that starting with a low steam rate gives better oil yield. This effect is indicated in Figure 1. When the steam rate R_{stm} , was increased from 0.5 to 1.4 [kg_{steam} / kg_{material} · h], the oil recovery Y_R , decrease from 80% to 45%, a 35% decrease in about two hours distillation time.

The same steam rate effect on the yield calculated by equation (7), can be seen in Figure 2. A high steam rate in the beginning of the steam distillation gives reduced oil recovery rates. In one hour distillation time, oil recovery rate R , decreased from 2.7×10^{-2} to 0.6×10^{-2} [L_{oil} / kg_{material} · h]. The effect can also be seen clearly in Figure 2 for shorter distillation times.

However, the steam rate should be increased gradually during the distillation time in order to achieve optimum oil yields. In Figure 1, the steam rate R_{stm} , ranging from 0.5 to 1.2 [kg_{steam} / kg_{material} · h] gives better overall recovery of oil Y_o .

Table 2. Composition of *Laurus nobilis* essential oil

| RRI* | Compound | % |
|------|---|------|
| 1032 | α -Pinene | 4.6 |
| 1076 | Camphene | 0.2 |
| 1118 | β -Pinene | 3.6 |
| 1132 | Sabinene | 7.8 |
| 1174 | Myrcene | 0.4 |
| 1177 | α -Terpinene | 0.5 |
| 1203 | Limonene | 0.8 |
| 1213 | 1,8-Cineole | 57.8 |
| 1255 | γ -Terpinene | 1.0 |
| 1280 | <i>p</i> -Cymene | 1.2 |
| 1290 | Terpinolene | 0.2 |
| 1553 | Linalool | 0.6 |
| 1565 | Linalyl acetate | t |
| 1612 | β -Caryophyllene | 0.2 |
| 1611 | Terpinen-4-ol | 2.2 |
| 1706 | Terpinyl acetate / α -Terpineol / Borneol | 8.3 |
| 2030 | Methyl eugenol | 0.2 |
| 2186 | Eugenol | 0.3 |
| | Monoterpene Hydrocarbons | 20.3 |
| | Oxygenated Monoterpenes | 67.9 |
| | Sesquiterpenes | 0.2 |
| | Others | 1.5 |

*: RRI Relative retention indices calculated against n-alkanes; t: trace (<0.1 %)

There are some important essential oils which occur well below the outer surface of the parent herb. These oils are found in many different parts of the plants. For Laurel leaves, the oils are found in between the palisade layer. In this case, liquid water plays significant part in bringing the subcutaneous oil to the herb surface. So the plant is classified as having “subcutaneous oils”. Diffusion takes place for recovery of these subcutaneous oils.

In dry plant materials, subcutaneous oils cannot easily be drawn to the surface of the material unless the material is wetted sufficiently enough for diffusion to take place. Therefore, for such materials low steam rates at the start of the distillation is required in order to sufficiently wet the plant material. Once the oils are surfaced, then steam rate can be increased step by step to evaporate the oil droplets by the passing steam. However, in fresh materials better yields can be obtained for subcutaneous oils if higher steam rates at the start of distillation are applied.

In the case of superficial oils meaning the oils contained in glandular hairs on the surface of the plant material such as Lamiaceae plants, again higher steam rates are expected to give better yields since diffusion for such oils is not important and they are easily released by breakage of the cuticular structure of the hairs and carried by the passing steam [2].

It was observed in all four experiments that in Laurel leaves, majority (80%) of the oil was recovered in a distillation time of two hours. The rest was recovered in the remaining two hours. Similarly, the recovery of 1,8-cineole, the main component, reached a maximum in the early fractions and gradually decreased in amount as the distillation proceeded. However, the recovery of the other main components, such as α -pinene, β -pinene, sabinene, γ -terpinene and the mixture of oxygenated

monoterpenes, (terpinyl acetate + α -terpineol + borneol) followed the completely opposite pattern as shown in the logarithmic chart of Figure 4.

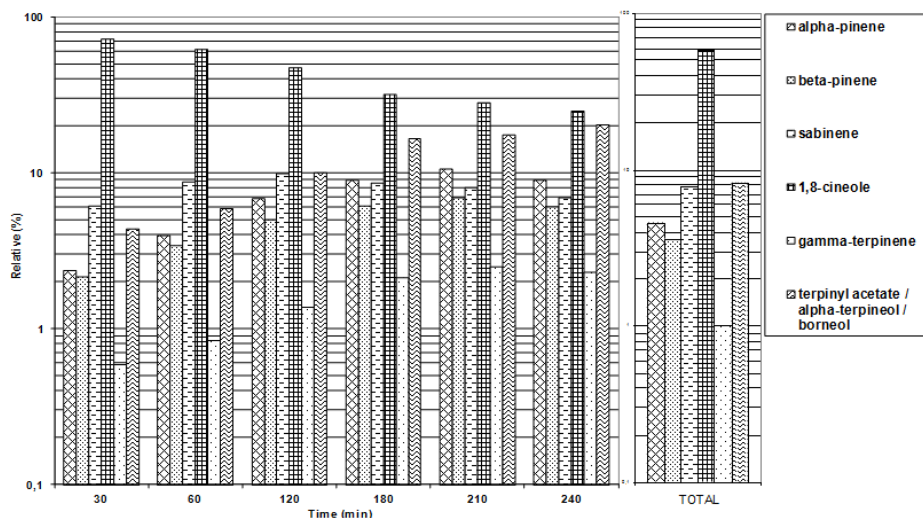


Figure 4. Distribution of the major components in differential oil samples

Optimum conditions for the pilot plant scale distillation of dried Laurel leaves are as follows:

1. Bed of material in the still should be well packed in order to prevent channeling.
2. Prior to distillation, the plant material should be wetted with water in order to facilitate diffusion.
3. It is advisable to start distillation at a low steam rate and gradually increasing it over a time period of distillation (*ca.* 0.5 to 1.5 [kg_{steam} / kg_{material} · h]).

Since 80% of the oil is recovered in the first two hours, distillation time should not exceed two hours for economical reasons.

Nomenclature

m : mass of moisture free basis plant material; [kg]

m_w : mass of wet plant material; [kg]

R : oil yield rate; [L_{oil} / kg_{material} h]

R_{sm} : steam rate; [kg_{steam} / kg_{material} h]

t : time; [h]

T_D : differential time; [h]

w : water (moisture) content of plant material; [kg]

V_c : cumulative oil volume; [L]

V_t : volume of oil recovered at time t; [L]

V_{it} : volume of oil recovered at infinite time; [L]

V_{OR} : overall relative oil in differential time;

V_T : volume of oil recovered at the end of distillation; [L]

Y_L : laboratory scale oil yield; [L_{oil} / kg_{material}]

Y_o : overall relative oil yield;

Y_P : pilot plant scale oil yield; [L_{oil} / kg_{material}]

Y_{Pt} : pilot plant scale oil yield at time t; [L_{oil} / kg_{material}]

Y_R : relative oil yield;

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