Optimization and Extrapolation to Pilot Scale of Essential Oil Extraction from *Pelargonium Graveolens*, by Steam Distillation

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Abstract

Essential oil extraction of *Pelargonium graveolens* that uses the extraction by steam distillation principle was studied at two scales: Laboratory scale and pilot scale. Firstly, a simple mathematical model was considered in order to optimize the effect of parameters that can influence the yield at the initial stage. This mathematical model is in the form of first polynomial degree with 3 variables for which the coefficient evaluation is done by multilinear regression. In order to reduce errors between calculated and experimental values, an optimal design was realized then followed by corresponding experiments to the plan. The complete factorial design ² was used. The obtained model is as shown: 

\[ Y = 2.156 - 0.183X_1 + 0.060X_2 - 0.038X_3 + 0.035X_1X_2 + 0.037X_1X_3 \]

Plant mass, extraction time and condensation output displayed interesting effect on extraction yield. Secondly, an empirical model was considered to extrapolate this result at different scales. The state of the gaseous fluid inside the conducting tube was taken into account. In order to get fluids with similar characteristics, the value of the charge loss was conserved within the two scales. Actually, the charge loss determines the balance state of the fluids in the tube. Then, this was combined with a kinetic model of first order, a law concordant well between experimental and calculated results was found.

Keywords: Essential oil; Steam distillation; *Pelargonium graveolens*; Model

Introduction

The geranium or *Pelargonium graveolens* is an aromatic plant highly used in cosmetics, in sanitary products and also in food preparation. The exploitation of natural products, like essential oils are privileged compared to synthetic products. The latter being more toxic often causes inauspicious secondary effects. In addition, these chemical products are some times less efficient than natural products. In developing countries like Madagascar, problems of process optimization are often forgotten. This leads to relatively high cost price of the finished products while labor cost remains very low. In spite of the permanent increase in demand for essential oils, extraction yield is low. For geranium, this yield is about 1.5‰ [1].

Few theoretical tools are currently available for studying scale change in the extraction process of essential oil that uses steam distillation principle.

A study on extraction yield improvement for *Pelargonium graveolens* essential oil using steam distillation and extrapolation of the results obtained from laboratory experiment to pilot scale. This yield noted Y is defined as:

\[ Y = \frac{\text{Mass of extracted oil}}{\text{Mass of plant}} \times 1000 \]  

(1)

The first step of the work consists in representing this Y function by one simple mathematical equation of the first order based on different parameters that influence this yield. These different variables were taken from in the literature. Experiments were planned according to a complete factorial plan ² In order to insure the quality of the obtained oil, a quality control by Gazeous Phase Chromatography [GPC] was undertaken and was compared with reference samples.

The second step allowed the extrapolation to the pilot scale by considering empirical appropriate equations. Actually, the steam passing through the tube is very important and defines the equilibrium state of the steam, consequently, the extraction yield [2].

Materials and Methods

Steam distillation principle

Steam distillation also called “water vapor distillation” or “hydro-distillation” permits to distillate without rectification volatile substances from crude products. The steam containing the oil is condensed in the serpentine before recuperation in the essential oil decantation container where the water and the oil are separated by gravity (Figure 1).

The study is done in two steps

The first step allows revealing the Y yield, here defined by a simple mathematical model based on different selected parameters. The identified factors that influence the yield are: the extracting time \( X_1 \), the condensation output \( X_2 \), the division factor of the plant material \( X_3 \), the relation between the plant mass and the introduced water mass \( X_4 \), the water loss by the plant material \( X_5 \) and the temperature \( X_6 \).

Pressure can also influence the yield. For this study, 3 variables were selected

\( X_1 \): The mass of introduced plant
\( X_2 \): The extracting time
\( X_3 \): The condensation output

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The least square constraint: \( E \) must be minimal.

The polynomial coefficients have a simple meaning: \( a_k \) gives the response function average, and indicates the effect of each parameter and \( a_k \) shows the interaction effects [3].

The coefficient estimation is done by multilinear regression. The principle is as following:

Let’s consider the model (2):

\[
Y = a_0 + a_1X_1 + a_2X_2 + \ldots + a_{n-1}X_{n-1} + a_nX_n + \ldots
\]

The second step allows associating the mathematical model with empirical equations in order to extrapolate the results to the pilot scale. The steam contained in the conducting tube can have some charge loss. A too low mass of plant is not recommended because the quantity of oil will be too small and this will influence profitability compared to the quantity of energy spent.

First step: Mathematical treatment

We will represent the extraction yield \( Y \) by a simple mathematical equation of this form:

\[
Y = a_0 + a_1X_1 + a_2X_2 + \ldots + a_{n-1}X_{n-1} + a_nX_n + \ldots
\]

The estimation of \( A \) is obtained through the resolution of the different factors on the response function. The factorial plan at 2 levels noted 2\(^k\) (K= number of variables) suits well our expectation in solving the problem.

The principle is to give to each factor two reduced centered levels noted -1 and +1. The lower level for each factor corresponds to -1 and the higher level to +1. In order to study the 3 variables (K=3), the complete factorial design presents 2\(^3\) = 8 lines and 3 columns for each factor.

The construction of the plan is done as following (Table 1):

First factor column: alternating -1 and +1

Second factor column: alternating -1 and +1, 2 by 2

| Test no | \( X_1 \) | \( X_2 \) | \( X_3 \) |
|---------|---------|---------|---------|
| no 1    | -1      | -1      | -1      |
| no 2    | +1      | +1      | +1      |
| no 3    | -1      | +1      | -1      |
| no 4    | +1      | +1      | -1      |
| no 5    | -1      | -1      | +1      |
| no 6    | +1      | -1      | +1      |
| no 7    | -1      | +1      | +1      |
| no 8    | +1      | +1      | +1      |

Table 1: Complete factorial design 2\(^3\).
Second step: Laboratory result extrapolation to the pilot scale

In order to preview the system behavior beyond the explored field, that is the pilot scale, we based our study on the similitude principle for which, [4] the relation between one or several parameters are the same for the two scales in order to reproduce the obtained results within a greater scale.

The principle is as following:

The steam coming out of the distillation balloon to the refrigerator passes through a conducting tube. In this tube, the steam might have some charge loss.

A too high charge loss may lead to partial damage of the main product and gives bad quality oil; a too low value leads to a premature condensation of the steam and consequently a lower extraction yield [5].

Therefore, to have the same fluid characteristics all the way through the trajectory in the conducting tube, we kept the value of the charge loss constant in the two scales;

The empirical expression of this charge loss "regular charge loss" is given by the equation:

\[ \frac{v^2}{2g} + z_1 + \frac{p_1}{\rho g} = \frac{v^2}{2g} + z_2 + \frac{p_2}{\rho g} + \Delta P \]  

(7)

If the steam encounters obstacles, it will have a particular charge loss which will add up to the regular charge loss. Its expression is given by (7):

\[ \Delta h = \frac{L \cdot v^2}{D_h \cdot 2g} \]  

(8)

The charge loss was determined by using the software MATLAB-SIMULINK [6]. Matlab helps to make simple and quick simulation and it permits to solve the problem efficiently.

During the calculation step, we have marked the fluids in the laboratory with * to keep the charge loss value constant and we get [2]:

\[ \Delta P^* = \Delta P \]

In our laboratory experiment we used a 2 liter balloon. The geometrical similitude allows keeping the relation plant mass \((m_p)_1\) / alembic volume \((V_{alembic})\) constant in the two scales. Thus we have:

\[ \frac{m_p}{V_{alembic}} = \frac{m_p^*}{V_{alembic}^*} \]  

(9)

This expression (8) allows calculating the introduced plant mass according to the alembic capacity that is used in the pilot unit in relation with the plant quantity put in the 2 liter balloon.

We also know that the number of the Reynolds's value \((R_e)\) determines the type of flow. When the \(R_e \) value is inferior to 2000, the flow is laminar [7].

This type of flow corresponds to our case. The Reynolds’s number [7] is expressed by the relation (10):

\[ R_e = \frac{\rho v D}{\mu} \]  

(10)

The charge loss coefficient for a laminar flow is given as follow:

\[ f = \frac{64}{R_e} \]  

(11)

As for the condensation output extrapolation, we used equations found in the case of an agitated reactor with discontinued run in liquid phases. In fact, this extraction method displays an analogy with that type of reactor. With the aid of some hypotheses, the characteristic speed equation is given by [8-9]:

\[ v_i = \frac{1}{v} \frac{dN}{dt} \]  

(12)

By introducing the rate of condensation, one gets:

\[ r = \frac{1}{V} N_o \int_0^t \frac{dx}{dt} \]  

(13)

The resting time to a given progress is:

\[ t = \frac{N_o}{V} \int_0^t \frac{dx}{r} \]  

(14)

\( V \) being the reactor volume, \( N_o \) the mole number at time \( t \).

The hypotheses are:

Heat repartition is considered homogenous in the two scales. The speed of the fluid remains constant during the experiment.

The reaction speed is described by the equation analogous to Van’t Hoof's and it expression is as follow [9]:

\[ v_i = K \cdot X \]  

(15)

\( [X] \) Represents the oil concentration at time \( t \) and \( p \) the reaction order. This concentration is the quantity of obtained oil per water volume unit distilling the oil.

- We consider that the flow is continual, that means that the elementary fluid mass that flows between the tube walls is the same. In this case the mass output is given by the relation [7]:

\[ q_m = \rho \cdot S \cdot v = \text{const} \cdot t \]  

(16)

Results

Mathematical treatment of the data

Confirmation of the parameters choices

a. Mass of introduced plant: The more plant one has in the same capacity balloons, the more difficult the oil at the bottom of the container can arrive on top surface in the same conditions. It is therefore possible that an important quantity of the raw material could cause burning and influence the quantity of oil obtained. This can lead to the reduction of the yield. This parameter is noted \( X_1 \).

b. Extraction time: Without considering the spent energy, a too long extraction time can cause burning of the prepared oil. In turn a too short operation time decreases the extraction yield because some part of the oil cannot be extracted from the plants. The extraction time noted \( X_2 \) varies from 3:30 h to 4:30 h.

c. Condensation output: We think that this factor is interesting between 3 and 6 ml/mn. It will be noted \( X_3 \). The condensation output determines the charge loss value. In fact, the charge loss is related to the square of the fluid speed.

Attribution of the variable levels

The experimental designs which are the translation of the reduced centered variables into real variables are given in table 2.

Experimental results

In order to estimate the experimental errors of the extraction yield,
we realized five automatized repetitions in the center of the field, and
the greatest difference compared to the experiment averages does not
 go over 3%. Each experiment imposed by the plan is repeated twice.
The design used is a complete factorial design $2^3$ (Table 3).

It gives the model according to the equation (4) that represents the
yield $Y$ in relation with the different parameters:

$$Y = 2.156 - 0.183X_1 + 0.060X_2 - 0.038X_3 - 0.012X_1X_2
+ 0.035X_1X_3 + 0.037X_2X_3$$ (17)

Where $Y$ is the yield directly expressed in % and the parameters
are the reduced centered coordinates between -1 and +1. Note that the
levels -1 and +1 correspond respectively to some real lower and higher
values of the parameters.

**Statistical results**

a. **Predicted coefficient - standard deviation:** The MODDE
software of UMETRI AB Society was used for the calculation.
The statistical study allowed to validate the model and to
describe the parameter effects on the field (Table 4).

b. **Regression significance:** Table 5 presents the statistical studies
carried on to evaluate the regression significance.

c. **Correlation matrix:** The correlation matrix is presented in the
following table 6.

d. **Individual significance of the coefficients:** The individual
significance of the coefficients is presented in table 7.

e. **Isoreponse curves:** Considering the $X_1$ factor constant to -1
value and the 2 parameters $X_1$ and $X_2$ taken as variables. In
these conditions the isoreponse curves take the form given in
figure 2.

**Study of the $Y$ response function**

a. **Statistical validation of the model:** The comparison of variance
$1/F$ with $F_{0.05,1,2}$ of SNEDECOR, reveals that the hypothesis of

Table 2: Variables levels.

| Test no | $X_1$ | $X_2$ | $X_3$ | Oil weight in g. | Yield in % |
|---------|-------|-------|-------|-----------------|------------|
| test no1 | -1    | -1    | -1    | 0.43            | 2.38       |
| test no2 | +1    | -1    | -1    | 0.44            | 2.43       |
| test no3 | -1    | +1    | -1    | 0.11            | 1.29       |
| test no4 | -1    | -1    | +1    | 0.79            | 2.38       |
| test no5 | +1    | +1    | -1    | 0.79            | 2.38       |
| test no6 | +1    | +1    | +1    | 0.79            | 2.38       |
| test no7 | 0     | 0     | 0     | 0.66            | 2.20       |
| test no9 | 0     | 0     | 0     | 0.66            | 2.20       |

**Table 3: Experimental results on centered reduced coordinates.**

| Test no | $X_1$ | $X_2$ | $X_3$ | Oil weight in g. | Yield in % |
|---------|-------|-------|-------|-----------------|------------|
| test no1 | -1    | -1    | -1    | 0.43            | 2.38       |
| test no2 | +1    | -1    | -1    | 0.44            | 2.43       |
| test no3 | -1    | +1    | -1    | 0.11            | 1.29       |
| test no4 | -1    | -1    | +1    | 0.79            | 2.38       |
| test no5 | +1    | +1    | -1    | 0.79            | 2.38       |
| test no6 | +1    | +1    | +1    | 0.79            | 2.38       |
| test no7 | 0     | 0     | 0     | 0.66            | 2.20       |
| test no9 | 0     | 0     | 0     | 0.66            | 2.20       |

The paragraph b representing the global signification of the model,
explains that the regression is significant. This means that the effect of
the parameters on the response is not negligible and that the model
describes well the phenomenon[6].

The correlation matrix symmetry (Table 6) indicates that the factors
are not correlated. Thus, the calculation of the multilinear progression
is valid for presenting the phenomenon[6].

b. **Interpretation display:** The relation (17) shows that the
different parameter coefficients are not very high. This
indicates that the parameter levels were well chosen because
the optimum should be very near to the experimental field [3].

The average quantity of extracted oil is 2.156%. This is higher
than the yield observed in the literature.

The mass of introduced plant has a negative effect on the yield.
This is expected: A more important quantity of plant material leads to
a lower yield because the oil at the bottom of the container has difficulty
to come up to the surface. The extraction time has a relatively low but
significant positive impact. Actually, for an extraction of 300 g of plants
under the same conditions as the experiments 1 and 5, the difference
in oil weight extracted is about 67 g. Its cost is equal to the labor for 3
extraction operations.

Table 4: Model coefficient and standard deviations.

| Terms | Coefficients | Standard deviation |
|-------|--------------|--------------------|
| Constant | 2.156 | 0.015 |
| $X_1$ | -0.183 | 0.017 |
| $X_2$ | 0.060 | 0.017 |
| $X_3$ | -0.038 | 0.017 |
| $X_1X_2$ | -0.012 | 0.017 |
| $X_1X_3$ | 0.035 | 0.017 |
| $X_2X_3$ | 0.037 | 0.017 |

Table 5: Signification de la régression.

| $X_1$ | $X_2$ | $X_3$ | $X_1X_2$ | $X_1X_3$ | $X_2X_3$ | Re |
|-------|-------|-------|----------|----------|----------|-----|
| 1     | 0     | 0     | 0        | 0        | 0        | -0.899 |
| 0     | 1     | 0     | 0        | 0        | 0        | 0.295 |
| 0     | 0     | 1     | 0        | 0        | 0        | 0.184 |
| 0     | 0     | 0     | 1        | 0        | 0        | 0.061 |
| 0     | 0     | 0     | 0        | 1        | 0        | 0.172 |
| 0     | 0     | 0     | 0        | 0        | 1        | 0.184 |
| Re    | -0.899 | 0.295 | -0.184 | -0.061  | 0.172    | 0.184 |

Table 6: Correlation Matrix.

| $X_1$ | $X_2$ | $X_3$ | $X_1X_2$ | $X_1X_3$ | $X_2X_3$ | Re |
|-------|-------|-------|----------|----------|----------|-----|
| 1     | 0.677 | 0.005 | 262.9    |          |          |     |
| 0.677 | 1     | 0.013 | 28.42    |          |          |     |
| 0.005 | 0.013 | 1     | 11.10    |          |          |     |
| 0.005 | 0.013 | 0.110 | 10.05    |          |          |     |
| 0.005 | 0.013 | 0.110 | 11.00    |          |          |     |

Table 7: Individual significance of the coefficients.
The isoresponse curves that the theoretical optimum is about 2.4 to 2.5 % corresponding to the operating conditions of \( X_1 = -1 \), \( X_2 = -1 \) et \( X_3 = +1 \); which is not the experimental case. In fact, the no.2 experiment attracts particularly our attention. The extraction time at the -1 level, therefore of 3:30. The plant mass is high and the condensation rate is relatively low. Despite the low yield of 1.95 %, less energy is spent to collect a sufficient quantity of oil. We consider these operating conditions as optimal.

Therefore, it is no sufficient to obtain some oil and to be satisfied from the studies on the effects of the different parameters. The oil quality must be controlled and compared with the standard oil quality.

**Quality control of the extracted oil**

The sample no.2 will be identified with its chemical composition and its physic –chemical characters in order to insure its quality according to the AFNOR norms. The physical characteristics such as the organoleptic characteristics, the relative density and the refractory index, show that the norms were followed.

To identify the constituents of our sample, we used the GPC based on the given operation conditions. The obtained chromatogram was compared with a standard chromatogram in order to identify the oil constituents. The identification of the chromatogram picks allows finding about 95.75% of all the constituents. The oil is of good quality and the rhodinol content (citronellol + geraniol) of 44% indicates that the oil is of Bourbon type [1].

**Extrapolation of the results to the pilot scale**

Our objective was to establish some extrapolation rules for industrial unit designs and at the same time study the system behavior outside the laboratory field. The studied field corresponds to 350 and 1000 Liter capacity alembics.

The relation (8) gives the extrapolation law of the introduced plant mass depending on the alembic capacity. For a 350l alembic, this mass varies respectively between -1 and +1 levels from 31 kg to 73 kg. For a 1000 l alembic capacity it varies from 90.5 kg to 209.5 kg.

Extraction time is an intensive value and does not depend on the quantity of plant material. At a greater scale, this time remains between 3:30 h and 4:30 h.

The relations (12), (15) and (16) allow determining the condensation output. For the 350l alembic, the recommended output varies from 21 to 63 l/h and from 60 to 180 l/h for a 1000 l alembic.

In order to obtain the same fluid characteristics throughout its trajectory, the conducting tube should be sized.

The relations (5), (6), (7), (10), (11) allow to calculate the size of the tube and to know the value of the charge loss that should not be passed. For a conducting tube of 13 cm diameter, the conducting tube length should be 2.30 m.

We have determined \( K_c \) and \( p \) from relations (12), (15) and (17) between 3:30 h and 4:30 h times:

\[
K_c = 0.027 \text{ h}^{-1} \text{ et } p = 1.
\]

Thus, the simulation of *Pelargonium graveolens* essential oil extraction was realized from the experiment n°4 at larger scale by associating two mathematical and kinetic models (Figure 3).

We note that the difference between the calculated and experimental values is relatively low (about 2%). At the end of the extraction, one notices a slight deviation between the model and the experimental values. This confirms on one hand that we are very near the optimum because the yield is tending to stabilization, and on the other hand, the chemical reaction phenomenon is less dominant than at the beginning of the operation [9].

In spite of the optimization and sizing study process complexity, the association of mathematical and simple kinetic models has simplified the simulation of the extrapolation of the *Pelargonium graveolens* essential oil extraction by correctly choosing the parameters to be studied. These results reveals that the proposed models simulate well the essential oil extraction going from the laboratory phase to the pilot scale extrapolation.
Conclusion

This extraction technique is often used in Africa to extract these plants oils. The geometrical similitude and the kinetic model (5),(6), (10), (11), (12), (15) and (17) that we elaborated allow displaying the kinetic model efficacy when it is associated with simple mathematical model. In fact, the chemical reaction phenomenon is present during the extraction that uses the steam distillation. The conducting tube sizing is necessary during the extrapolation step. Former studies on material transfer model during essential oil extraction from coriander fruits confirm this thesis on the presence of the chemical reaction [8].

In addition, experimental studies allowed improving the yield at pilot scale and bringing an extrapolation theory of essential oil extraction that uses this steam distillation principle.

- Importance of the charge loss value that permits the sizing of the conducting tube,
- Presence of the chemical reaction during the extraction, consequently, the speed general equation of the reactions allows extrapolating the condensation output.

Nomenclatures

| Symbol | Description |
|--------|-------------|
| Y      | Extraction yield (%a) |
| GPC    | Gazeous phase chromatography |
| $\Delta h$ | Particular charge loss |
| $\Delta P$ | Regular charge loss |
| Re     | Reynold’s number |
| $\rho$ | Volume mass of gaseous fluid |

References

1. Manda Rabe Ravelona (1995) Contribution to the project of géraniculture to Ambatolampy. AGRIKA Memory Graduation.
2. J.LIETO (1998) Chemical engineering at the use of chemical. Lavoisier Technique et documentation.
3. Silou T, Malanda M, Loubaki L (2004) Optimization of the extraction of the essential oil of Cymbopogon citratus with a full factorial design 2$^3$. J Food Eng 65 : 219-223.
4. Phillipson Robert RANDRIAMIARISOA (1995) Quick Guide and useful for producing essential oil.
5. Kirk Othmer concise Encyclopedia of chemical technologie.
6. SADO G, SADO MC (2000) Design of experiments: the experimental quality assurance. AFNOR, Paris.
7. Desjardins D, Combarious M, Bonneton N (2006) Fluid Mechanics. Dunod.
8. Martial Chabanel (2011) Thermodynamique chimique. Ellipses Marketing.
9. Benyousef EH, Hasnî S, Belabbes R, Bessiere JM (2002) Modelling of mass transfer during extraction of the essential oil of coriander fruits. Chem Eng J 85 : 1-5.