

# STEAM EXTRACTION OF ESSENTIAL OILS: INVESTIGATION OF PROCESS PARAMETERS

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The steam extraction was used to investigate the process parameters important in the extraction of essential oils from the leaves of the Eucalyptus tree. The diffusion process and the application of Fick's law, as well as the temperature dependence of the diffusion coefficient were exhibited. This has enabled the setting up of a model that takes account of the initial amount of oil in the leaves and the effect of temperature. From the literature, it can be seen that the oil collected after an extended time represents all the oil that is present in the material initially. The research conducted indicates that the extraction temperature has a significant effect on the percentage of the initial oil that is eventually extracted, with the amount of oil extracted apparently decreasing for lower temperature. This effect could, however, be modelled accurately by simply considering the temperature dependence of the diffusion coefficient. This model can lead to the design of an optimum steam extraction plant.

L'extraction par vapeur a été utilisée pour étudier les paramètres importants du processus de l'extraction des huiles essentielles des feuilles de l'eucalyptus. Le processus de diffusion et l'application de la loi de Fick, ainsi que la dépendance par rapport à la température du coefficient de diffusion ont été montrés. Ceci a permis la mise en place d'un modèle qui prend en compte la quantité initiale d'huile dans les feuilles et l'effet de la température. Le coefficient de diffusion établi dans cette étude de recherche tombe dans l'intervalle des valeurs rapportées par la littérature pour les huiles essentielles. À partir de la littérature, on peut voir que l'huile collectée après une période prolongée représente la totalité de l'huile présente initialement dans le matériau. L'étude présente indique que la température d'extraction a un effet significatif sur le pourcentage de la quantité initiale d'huile éventuellement extraite, la quantité extraite diminuant apparemment pour les températures plus basses. Cet effet pourrait cependant être modélisé exactement en considérant simplement la dépendance par rapport à la température du coefficient de diffusion. Ce modèle peut conduire à la conception d'une usine d'extraction optimale à la vapeur.

**Keywords:** Eucalyptus essential oil, steam extraction, diffusion, mathematical model

## INTRODUCTION

Essential oils are volatile oils, generally odourous, which occur in certain plants or specified parts of plants, and are recovered by accepted procedures, such that the nature and composition of the product is, as nearly as practicable, unchanged by such procedures.

This is an important definition. It specifies clearly that the nature and composition of the oil must be unchanged by the process of extraction, which is one reason why steam is an appropriate method of extraction. Furthermore, because steam distillation has been the extraction method for most essential oils, the market accepts steam-distilled oil as normal oil. Oil derived by another technique might be of slightly different chemical

composition and therefore might not be accepted by the market as normal oil (ISO, 1968).

Essential oils are used widely by the pharmaceutical and cosmetic/perfumery industries as well as in aromatherapy and alternative medicine.

Essential oils have some distinctive characteristics, which make them a very valuable commodity with many industrial uses and applications. Their aromatic value enables them to be used as

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flavourings in both the food and beverage industries. These oils are also widely used in both the cosmetic and pharmaceutical industries (Worwood, 1990).

With such applications, there is a huge demand for essential oils worldwide and hence they have been traded internationally for several centuries. There is, hence, a need to improve the quality and quantity of the essential oils produced as they have a very competitive and profitable market worldwide (Learmonth and Westhuizen, 2002).

The chemical composition of the essential oils is important in determining their quality and consequently price in the market (Learmonth and Westhuizen, 2002). It is therefore important to note and understand some of the parameters such as temperature, pressure, and time of extraction may affect the quality and yield of essential oil (Guenther, 1960).

Essential oils can be extracted using a variety of methods, although some are not commonly used today. Currently, the most popular method of extraction is steam extraction, but as technological advances are made more efficient and economical methods are being developed. These include methods such as solvent extraction, supercritical fluid extraction, cold pressing, and microwave extraction. The suitability of extraction method varies from plant to plant and there are significant differences in the capital and operational costs associated (Sheridan, 2000). In steam extraction, there is a dearth of information regarding the effect of temperature on diffusion coefficients, and this is needed in the design and operation of extraction units.

This project seeks to facilitate import substitution by enabling the development of small-scale process plants to extract essential oils from local plants using steam extraction.

## EXPERIMENTS

The essential oils were extracted using the fresh leaves of trees (*Eucalyptus grandis*) collected from a single location in a reserved garden in the Region 9 area of Johannesburg (South Africa).

Samples with a mass of approximately 20 kg were prepared by loading on an electronic balance. The plant material from a given sample was packed into the extraction chamber so that distillation could commence. Proper charging is very important; otherwise the steam channels through the plant material and low yield results.

The first load was used to set-up and establish the procedure and to determine processing parameters. The experimental equipment of steam extraction of essential oils is illustrated schematically in Figure 1 below.

Steam was generated using a 15 kW boiler, capable of producing 15 kg steam/h, connected to three-phase power. Galvanized tubing (12.5 mm), fitted with an outlet and steam flow control valve, was used to transfer steam to the base of a stainless steel extraction chamber. The extraction chamber, a large pot with a stainless steel jacket sealed with a rubber O-ring and clips, was 60 cm in diameter and 70 cm high.

The sample was placed in a mesh basket, which was positioned in a slightly elevated position in the extraction chamber. After passing through the plant material, steam was released through the top of the jacket into a condenser coil, constructed from 32 mm stainless steel tubing. The extraction chamber was connected to the cooling coil via a Quick coupling. The condenser coil was immersed in a stainless steel drum, which was cooled by passing running water, with a hose, through the drum. An outlet, at the top of the drum, allowed the hottest water to run out to be replaced by cooler water. A second outlet, at the base of the cooling drum, was used to drain water from the drum when necessary.

The distillate, at a flow rate of approximately 100 mL/min, was collected using 250 mL separating funnels. The batch was discontinued once the flow ceased. It is important to note that when the pressure reached the respective desired operating conditions, stripping was commenced by opening the steam flow valve which released the steam to flow into the steam line and eventually to the extraction chamber. The exit line of the condenser was observed for the first condensate drop in the

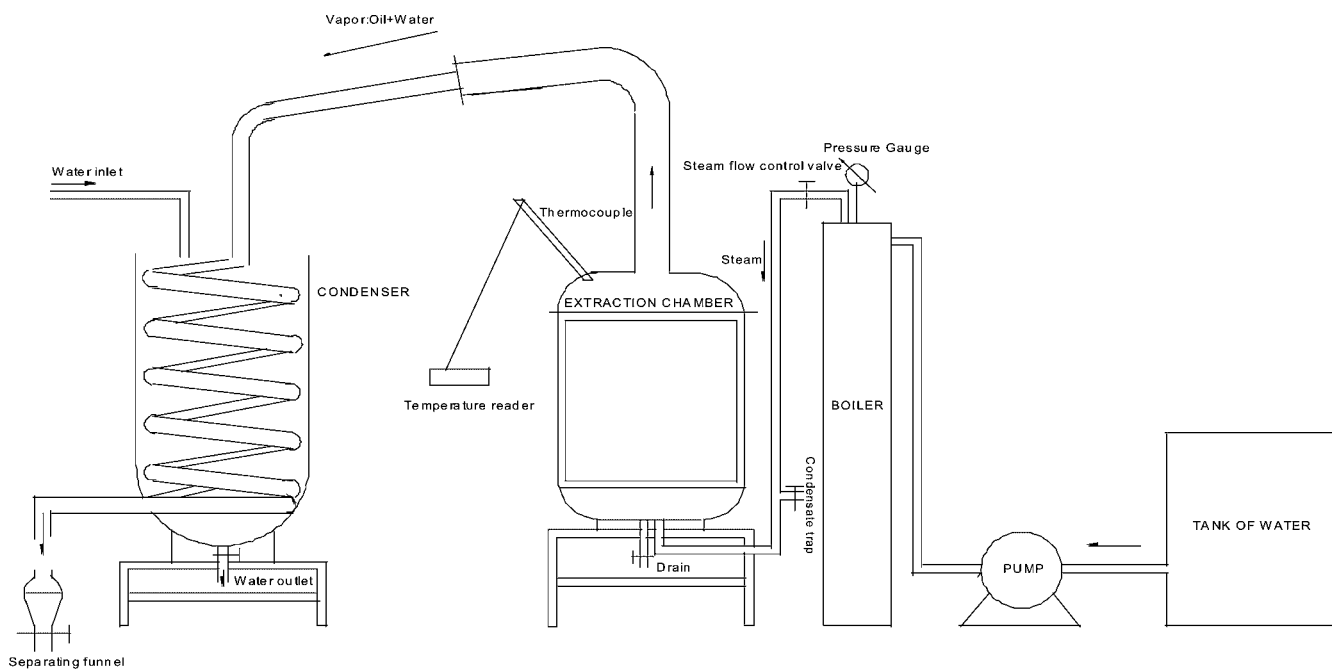


Figure 1. Schematic representation of the steam extraction equipment.

collector help on position, at which point a stopwatch was started. The time the first drop was collected was taken to be the initial extraction time of the oil extracts from the plant material.

Oil samples obtained during this study were stored in a dark glass bottles with suitable caps in cool place away from light.

The quantity of the Eucalyptus essential oil extracts was determined by use of an XP-300 top loading electronic analytic balance, which determined the mass to within accuracy of 0.001 g.

The composition of the Eucalyptus essential oils extracts was analysed using a CP-3800 Varian gas chromatograph (GC) unit. Gas chromatography involves a sample being injected and vaporised onto the head of a chromatograph column.

The gas chromatograph was filled with a fused silica capillary column, split-less injector, a vapour sampling loop and a Flame Ionisation detector (FDI). Chemically inert Helium gas was used as both a reference and carrier gas during sample analysis.

A sample of 2.0  $\mu\text{L}$  of each of the oil extract samples collected at different pressures and 100  $\mu\text{L}$  of Hexane (vaporised solvent) was prepared as dilute solution due to the sensitivity of the detection methods.

## RESULTS AND DISCUSSION

### Mathematical modelling

The Fick's law of diffusion was used because it is the more fundamental, straight forward way to model diffusion processes and can be used to solve for the diffusion coefficient  $D$ . It is assumed that the oil has mostly the same composition so that a single diffusion coefficient will suffice in the modelling.

A numerical method needed to be developed to describe the oil extraction process and mechanism during the steam extraction of Eucalyptus leaves. This model was used to fit the experimental data for the variation of oil leached with time and hence compare the diffusion coefficients of the oil extraction for different temperatures. The leaf was divided into sections of thickness  $\Delta x$ . If  $\Delta C$  is the concentration difference between two sections, Fick's first law (the diffusion flux along direction  $x$  is proportional to the concentration gradient) may be written as:

$$J = -D \frac{\Delta C}{\Delta x} \quad (1)$$

where  $J$  is the diffusion flux,  $D$  the diffusion coefficient,  $\Delta C/\Delta x$  the concentration gradient, and the minus sign in the equation means that diffusion is down the concentration gradient.

$$J = \frac{\Delta n}{A \Delta t} \quad (2)$$

From (1) and (2):

$$\frac{\Delta n}{A \Delta t} = -D \frac{\Delta C}{\Delta x} \quad (3)$$

After a short time  $\Delta t$ , the difference in the number of moles diffusing out and in a given section, respectively, are given by:

$$\Delta n_{\text{out}} = -A \Delta t D \frac{\Delta C_1}{\Delta x} \quad (4)$$

$$\Delta n_{\text{in}} = -A \Delta t D \frac{\Delta C_2}{\Delta x} \quad (5)$$

where  $\Delta C_1$  is the concentration difference in the section of the leaf and the next section in the direction of the edge of the leaf,  $\Delta C_2$  the concentration difference in the previous section of the leaf and the current section,  $\Delta n_{\text{out}}$  the number of atoms diffusing out of the leaf,  $\Delta n_{\text{in}}$  the number of atoms diffusing into the leaf during time interval  $\Delta t$ .

Net loss of  $n$  (the number of atoms diffusing through the area of leaf):

$$\Delta n_{\text{out}} - \Delta n_{\text{in}} = A \frac{\Delta t}{\Delta x} D (\Delta C_1 - \Delta C_2) \quad (6)$$

$$\frac{\Delta n_{\text{out}}}{A \Delta x} - \frac{\Delta n_{\text{in}}}{A \Delta x} = -D \Delta t \frac{(\Delta C_1 - \Delta C_2)}{\Delta x^2} \quad (7)$$

Drop in concentration:

$$C_{\text{out}} - C_{\text{in}} = -\frac{D \Delta t}{(\Delta x)^2} (\Delta C_1 - \Delta C_2) \quad (8)$$

$$C = C_0 - \frac{D \Delta t}{(\Delta x)^2} (\Delta C_1 - \Delta C_2) \quad (9)$$

From this equation, we can generalise:

$$C_n = C_{n-1} - D \frac{\Delta t}{\Delta x^2} (\Delta C_1 - \Delta C_2) \quad (10)$$

Hence one can calculate the concentration in a given section of leaf ( $C_n$ ) after a time  $\Delta t$  has elapsed using data from the previous time interval.

The model is very important in the scale-up process from laboratory work to an actual industrial process as well as providing an insight into the extraction mechanism (Bulley et al., 1993). The model development is represented by a simple schematic diagram (Figure 2) and the steps are listed below:

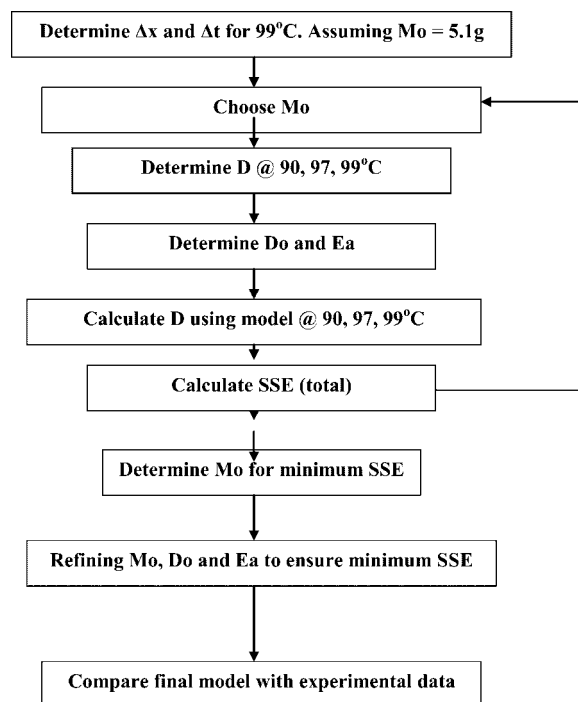


Figure 2. Schematic diagram of modelling steps.

The determining of  $\Delta x$  and  $\Delta t$  for 99°C was done by assuming the initial oil mass ( $M_0 = 5.1$  g). In order to determine the required number of segments (determined by the value of  $\Delta x$ ), the number of segments in the spreadsheet were increased until the best-fit value for  $D$  reached a steady value. It was then deduced that increasing the number of segments above 20 did not make the modelling more accurate. The same principle was followed to determine  $\Delta t$ . Here  $\Delta t$  was decreased until the best-fit value for  $D$  became constant, which happened when  $\Delta t$  was 0.30 min.

Equation:  $C_n = C_{n-1} - D \frac{\Delta t}{\Delta x^2} (\Delta C_1 - \Delta C_2)$  was used in the Microsoft Excel spreadsheet. We assumed that the percentage of oil condensate concentration at edge of the leaves to be equal to 0 because weight fraction of oil =  $(\frac{n_{oil}}{n_{H_2O} + n_{oil}})_{vapour} = (\frac{n_{oil}}{n_{H_2O} + n_{oil}})_{condensate} = 0.000011$  as calculated from amount of oil and water collected after condensation. This value can be seen to be close to 0. This since mass of oil and water collected are 5.1 and 60 000 g, respectively.

$$D = D_0 e^{-E_a/RT} \quad (11)$$

Equation (11) was also used in spreadsheet to determine the value of  $M_0$  (initial mass). From this value (5.1 g), the diffusion coefficient at different temperature was determined.

The best-fit values of  $D$  were then determined for each temperature and the SSE was calculated. In the model, the parameter  $D$  was directly adjusted using Equation (11) with the data from the spreadsheet. This parameter was estimated by minimisation of the sum of square of errors (SSE) between the experimental data obtained and the prediction from model.

The diffusion coefficients at 90, 97, and 99°C were employed to determine  $D_0$  and  $E_a$  using the linear regression as illustrate in Figure 3. Here Equation (11) was used in its logarithmic form:  $\ln D = \ln D_0 - (E_a/RT)$ . The best-fit values (minimising the SSE) of  $D$  from the spreadsheet for the various temperatures were used to calculate the  $\ln D$  values.

$$E_a = 135.8582021 \text{ kJ/mol}; D_0 = 102825214.7 \text{ m}^2/\text{s}.$$

The determination of diffusion coefficients using the model at 90, 97, and 99°C was done on the spreadsheet by fitting the experimental data. The best-fit values for the diffusion coefficient at the various temperatures were determined in Table 1.

The SSE was obtained from the spreadsheet by adding the SSE of all the data points for a given temperature in Table 2.

Several values of  $M_0$  were investigated and the one (5.4 g) that gave the smallest overall SSE was selected.

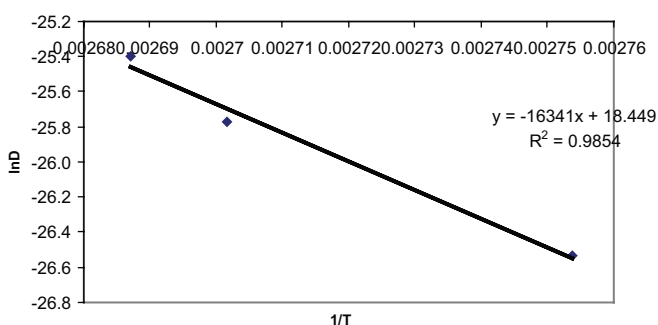


Figure 3. Determination of the activation energy:  $\ln D$  versus  $1/T$ .

Table 1. The best-fit values of diffusion coefficient  $D$

Minimum SSE	Diffusion coefficient ( $\text{m}^2/\text{s}$ )	Temperature ( $^{\circ}\text{C}$ )
0.364	$2.93000 \times 10^{-12}$	90
0.301	$6.86089 \times 10^{-12}$	97
0.404	$8.69860 \times 10^{-12}$	99

Table 2.  $D$  values versus temperature using the determined values of  $D_0$  and  $E_a$

Minimum SSE	Diffusion coefficient ( $\text{m}^2/\text{s}$ )	Temperature ( $^{\circ}\text{C}$ )
0.382	$2.94 \times 10^{-12}$	90
0.411	$6.89 \times 10^{-12}$	97
0.511	$8.74 \times 10^{-12}$	99

The refining of  $M_0$ ,  $D_0$ , and  $E_a$  was obtained by adding and minimising the SSE for all experiments (90 + 97 + 99°C).

From Figure 4, it can be seen that the values for  $D$  calculated from the final model (using Equation (11)) give a good fit to the experimental data, and this was only slightly less accurate than using the individual best-fit values determined for each temperature.

The final model was determined from the refining the values of  $D_0$ ,  $M_0$ , and  $E_a$  in Equation (11).

Here  $D_0$ ,  $M_0$ , and  $E_a$  were adjusted until the SSE for all the experiments was at a minimum.

From Figure 5 above, the optimum values of  $D_0$ ,  $M_0$ , and  $E_a$  were confirmed by minimising the SSE. Only small changes to the constants were required to truly minimise the SSE. It can be noted that the optimum value of  $E_a$  is much more accurately determined than that of  $M_0$  and  $D_0$ .

The best-fit adjusted parameter of the model (Diffusion coefficient) was in agreement with available literature data in terms of order of magnitude, backing up the model used.

The Figures 6–8 show the oil profiles in cross-section during leaching using the final model at the different temperatures used. Comparison of the figures indicates that oil is depleted faster at the higher temperature.

The resulting expression for the diffusion coefficient as a function of temperature can now be used to model the large-scale extraction of the essential oils from Eucalyptus leaves.

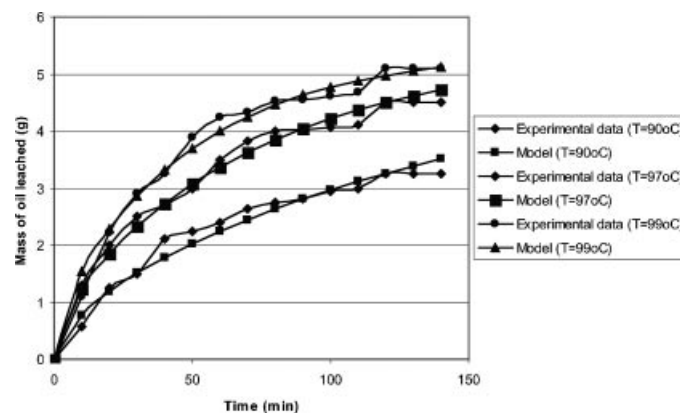


Figure 4. Fitting the theoretical oil extraction model to experimental data using final model values.



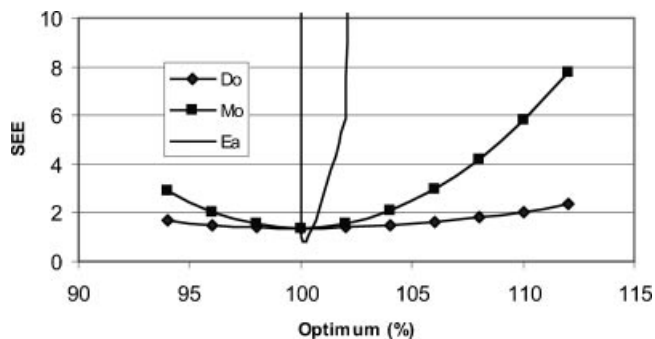


Figure 5. Optimization of  $D_0$ ,  $M_0$ , and  $E_a$  versus SSE.

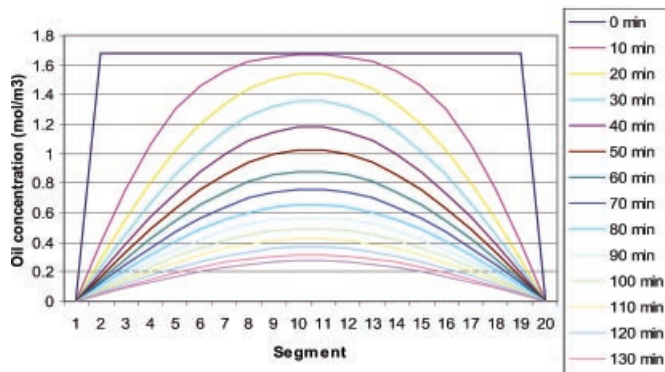


Figure 6. Oil profiles in leaf cross-section during leaching using final model at 90°C.

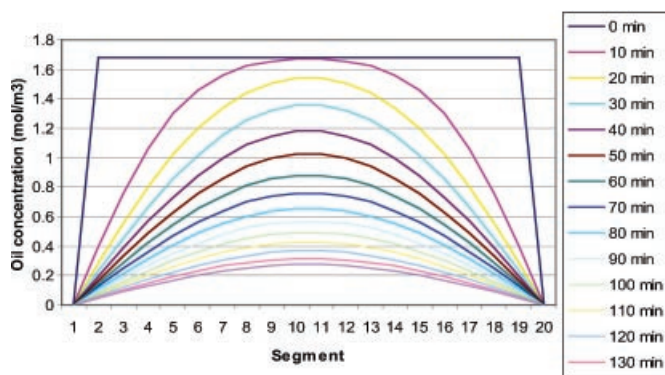


Figure 7. Oil profiles in leaf cross-section during leaching using final model at 97°C.

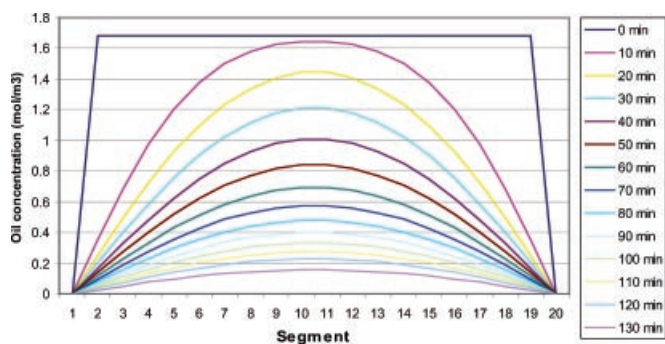


Figure 8. Oil profiles in leaf cross-section during leaching using final model at 99°C.

## CONCLUSIONS

This study followed an inter-disciplinary approach to investigate the effects of different operating parameters such as temperature and extraction time on the yield and quality of the Eucalyptus oil extracts obtained using steam distillation.

It was shown that an increase in temperature increases the extraction rate.

A mathematical model based on diffusion of essential oil in leaves was set up. Using a numerical method the best diffusion coefficient was established for different operating conditions by comparing the model concentration of oil leached in the leaves with the experimental amount of oil recovered; hence minimising the sum of squared errors.

The model parameter evaluation, attained in this work to laboratory-scale, could be useful during the scale-up of the extraction process and/or during industrial operation to evaluate the extraction time required to obtain a given yield, as confirmed in this study.

It was possible to verify the importance of the process parameters in the yield of the Eucalyptus essential oil by steam distillation by using a model to determine the concentration profiles at any time in the leaves during the extraction.

The experimental data for Eucalyptus essential oil yields were well correlated by the diffusion model. The modelling revealed that the assumption usually made that all the oil is removed during an experimental is not correct, as the amount of oil leached depends on the temperature of the test. Hence it is suggested that the model parameter evaluation, attained in this work on a laboratory-scale, could be useful during the scale-up of the extraction process and/or during pilot or industrial operation to evaluate the extraction conditions required to obtain a given yield.

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