

Article

Mathematical Model for the Removal of Essential Oil Constituents during Steam Distillation Extraction

Sharad Maharaj * and David McGaw

Chemical Engineering Department, The University of the West Indies, St. Augustine, Trinidad and Tobago; drmcgaw@gmail.com

* Correspondence: Sharad.Maharaj@sta.uwi.edu; Tel.: +1-868-662-2002 (ext. 83073)

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Abstract: Steam distillation is the conventional means by which oils are extracted in the flavour and fragrance industry. A mathematical model for the steam distillation extraction (SDE) of air-dried *Ocimum basilicum* (basil) leaves has been developed and tested using a small-scale pilot plant. The model predicts the removal of oil components from the plant matrix and subsequent transfer to the steam. It also accounts for the diffusional transfer of components within the leaf and the simultaneous convective transfer into the vapour phase while also respecting the individual components' volatilities. It has been applied vertically on an element-by-element basis through the bed for a mixture of major and minor components. The proposed SDE model appears to be a good match between predicted values and the experimental data. The model predicts a faster initial extraction rate for components such as α -pinene and α -terpinene, possibly due to preferential extraction of light, volatile components present in larger quantities.

Keywords: convective mass transfer; element-by-element; essential oil; extraction curve; *Ocimum basilicum*; steam distillation

1. Introduction

1.1. The Essential Oils Industry

Many plant species secrete complex chemicals that are extracted and incorporated into various consumer products, which may be classified under the following headings:

Food Flavours

Aromatherapy/Personal Care Products

Nutraceuticals/Pharmaceuticals

Perfumes

Various other uses, for example, insecticides, security sprays, dyes, and colourings

There are two main types of extracts, essential oils and oleoresins. Essential oils are the volatile oils giving rise to the odour from the plant with oleoresins having a much lower vapour pressure, boiling at higher temperatures.

Growing consumer preference for natural products has led to the development of novel applications in personal care and beauty products. In addition, demand in the medical industry, on account of growing consumer awareness regarding health benefits, and with the advantage of negligible side effects associated with the use of essential oils, is expected to spur increased applications. Grand View Research estimated the global essential oil market to be ~US\$5.5b in 2014 [1]. They stated that increasing essential oil penetration in aromatherapy, coupled with rising demand for fragrances and flavours, is expected to continue as a driving factor for the global market. Furthermore, Grand View Research estimated the United States essential oil market would double from ~US\$2.0b to almost ~US\$4.0b

in 2022 with breakdowns being given for the top 10 oils [1]. The European market was valued at ~US\$2.4b, but not expected to grow at the same rate [1].

1.2. Essential Oil Extraction Technologies

Steam distillation is the conventional means by which oils are extracted in the flavour and fragrance industry and for more than a century has been the most widely accepted process for distilling oils on a large scale. The SDE process consists of passing steam through a vertically oriented packed bed of the material from which the oils are to be extracted. A typical system is shown in Figure 1, which is a diagram of the specific unit used in the experimental programme carried out in this work. The steam acts both as a heating agent and carrier for the components extracted from the plant charge. The vapours exiting the extraction chamber are then condensed into oil and water. The extracted oil floats on the aqueous extract and can be collected when cooled as can be seen below in Figure 1. There are hundreds of SDE plants globally, currently producing the bulk of the US\$5.5b market in essential oils. The vast majority of these plants are located in rural areas of developing countries.

The more modern alternative to steam distillation is supercritical fluid extraction, whereby carbon dioxide at high pressure (above the critical point of ~73.8 bar and 31.1 °C) is passed through the bed of material. The capital cost is significantly higher than SDE; it requires a source of carbon dioxide and extracts the essential oils in slightly different ratios.

The work in this paper is geared towards attaining a better understanding of the Steam Distillation Extraction (SDE) process through reviewing SDE modelling, as presented in the literature. This involves identifying their weaknesses and introducing a new, more comprehensive model which is tested experimentally.

1.3. Published SDE Theories

Guenther has presented a detailed background to the process and extensive use of SDE in the production of essential oils. In contrast, more comprehensive theoretical analyses on modelling the process have been more recent [2].

Thus, a kinetic model was proposed by Romdhane et al. for the SDE of aniseed. The model was developed by treating the oil as if it was comprised of only one constituent and was solved by fitting the experimental results to the kinetic equation for an overall solid-steam mass transfer coefficient [3]. Kinetic models, however, are inherently limited in that their adjustable parameters have no physical meaning [4].

Cassel modelled the distillation of citronella oil from leaves and twigs based on Fick's law of diffusion. An effective diffusivity (D) was obtained by adjusting its value in the proposed model to fit the experimental data [5]. Cassel also applied the same model to basil leaves in another investigation [6]. In both cases, the individual constituents making up the oil and their respective volatilities were not considered. Instead, the oil was considered to be homogenous and constant for all particles making up the extraction matrix.

In the study published by Sovova which focused on the hydrodistillation of essential oils, she recognized that further model developments should incorporate the different volatilities of essential oil components, which would bring about changes in the distilled oil's composition during the extraction process. Sovova's study is the only one to date, which recognizes that all essential oils are a mixture of several components with differing volatilities. However, there was no further development of such a model [7].

More recent modelling by Xavier et al. considered the oil as a single component [8].

A review of kinetic models for essential oil extraction by Meziane et al. confirmed that to date, the predictive models simulate the transfer of a single component considering the solid phase resistance primarily while neglecting the essential oil concentration in the vapour phase [9].

1.4. Objectives of Research

The models identified in the literature survey are inadequate in that they:

1. Treat the packed bed as a fully mixed entity, whereas in operations involving passing gas through a fixed packed bed of particles, there will be vertical variations in both the gas and the particles in the bed.
2. Do not incorporate all of the resistances to transfer in their models.
3. Do not recognize that the essential oil separated from the particles is a mixture of components of differing volatilities.

An essential oil's monetary value depends on its final composition and any mathematical model attempting to predict the extraction of the oil must consider all the important quality components in the mixture, both major and minor. The work described in this paper presents a new model which recognizes the inadequacies of the reported models and shows how well the new model fits the experimental data obtained from the operation of a small pilot plant scale SDE unit using locally grown basil leaves.

2. Mathematical Model

Reference is made to Figure 1, which shows the small scale pilot plant SDE unit used to generate the experimental results. In Figure 1, it is seen that the extraction chamber was located within a stainless-steel chamber, through which steam was passed to provide a steam jacket for the extractor and significantly minimized heat losses during steam distillation. Since the equipment design provided adiabatic conditions, only the component transfer mechanisms need to be considered. As we shall show, our postulated SDE model acknowledges the essential oil as a mixture of different organic components and the transfer of individual oil components is considered.

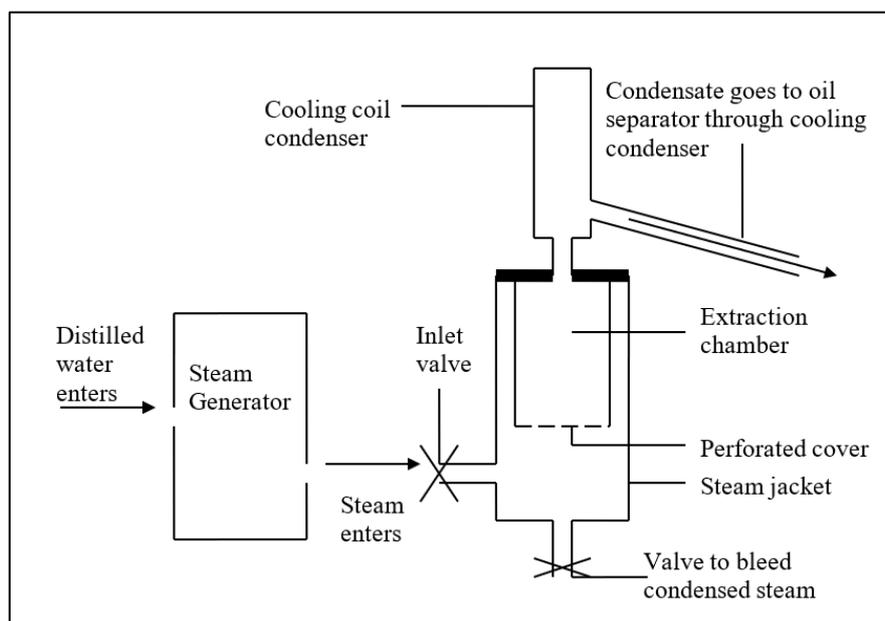


Figure 1. A schematic flow chart of the Steam Distillation Extraction (SDE) pilot plant apparatus.

2.1. Mechanisms of Component Transfer

There is normally a pre-treatment process of natural air-drying of the plant material, which, when coupled with the temperatures and pressures involved in SDE, results in a breakdown of cell walls within the material. In the case of the extraction of the essential oil from leaf materials, this heating and pressure allow for component diffusion to the adaxial (upper leaf surface) and the abaxial (lower

leaf surface) sides of the leaf. The oil sacs in *Ocimum basilicum* (basil) are located primarily in the subcuticular areas of the leaf. When steam is passed over the leaf surface, the leaf warms, and oil that has reached the point of vaporization diffuses to the surface and contacts the steam.

Considering the plant charge in terms of a series of vertical elements or layers, the concurrent component transfer operations that take place from the leaf to the steam are represented in Figure 2 below. The model proposes that there are three basic mechanisms relating to the transfer of oil from within the material to the steam passing through the bed as follows:

1. Transfer of the oil to the surface of the material by diffusion
2. Vaporization of the oil at the surface
3. Transfer of the oil from the surface of the material to the steam by convection

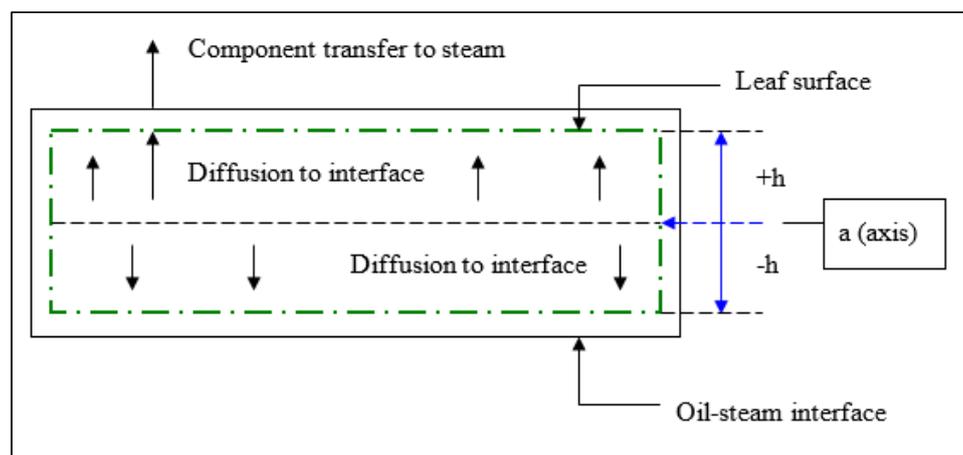


Figure 2. Component transfer mechanisms from leaf to steam.

The relevant equations for each of these are as follows:

2.2. Diffusion

To model the component diffusion from a leaf, we applied the diffusion equation derived by Crank for diffusion from an axis bounded by two parallel sheets [10].

Denoting x as the concentration of diffusing component that moves to the surface of the leaf over contact time, t and x_0 as the total concentration of the component in the leaf, the following relationship as developed is appropriate, as described in Reference [10]:

$$\frac{x}{x_0} = 1 - \frac{8}{\pi^2} \sum_{n=0}^{\infty} \frac{1}{(2n+1)^2} \exp\left\{-\frac{D(2n+1)^2\pi^2t}{H^2}\right\} \quad (1)$$

where, D = diffusion coefficient, m^2/s

$H = 2h$ = leaf thickness.

x = concentration of component in liquid mixture

2.3. Equilibrium Relationship

When each of the components initially reaches the surface of the particle/leaf, it will exert its vapour pressure at the temperature of the surface, assumed to be 100°C for steam distillation at around atmospheric pressure. Thus, the partial pressure of each of the individual components making up the oil at the surface can be determined using the following equation:

$$p^* = x(P), \quad (2)$$

where, p^* = equilibrium partial pressure, bar

P = vapour pressure of the pure component under the same temperature conditions, bar

x = concentration of component in liquid mixture

2.4. Resistance to Convective Transfer

Each component is then transferred into the bulk steam phase by convective mass transfer from the surface of the leaf. The following relationship can be used, based on the concept of a boundary providing resistance to component transfer from the charge surface, where components are at the point of vaporization, to the region of mixed vapour:

$$\frac{dp}{dt} = \frac{kA(p^* - p)}{V} \quad (3)$$

where, p = partial pressure of the solute in the free stream, steam, at time, t

p^* = equilibrium partial pressure that the individual component exerts at the boundary

k = convective resistance, m/s

A = leaf-steam interfacial area, m^2

V = volume of steam surrounding the particles in the bed, m^3

If A and V can be assumed constant, the time taken for the partial pressure of a component to increase from an initial value partial pressure, p_0 to a final value partial pressure, p , can be found by integration to give:

$$p = p^* \left(1 - e^{-\frac{kAt}{V}} \right) \quad (4)$$

The equation can be applied to determine the partial pressure of a component in the steam provided that values of p^* and k can be determined.

Incorporating this relationship into Equation (1) derived for p and substituting gives:

$$p = P x_0 \left(1 - e^{-\frac{kAt}{V}} \right) \left\{ 1 - \frac{8}{\pi^2} \left[\exp\left(\frac{-D\pi^2 t}{H^2}\right) + \frac{1}{9} \exp\left(\frac{-9D\pi^2 t}{H^2}\right) + \frac{1}{25} \exp\left(\frac{-25D\pi^2 t}{H^2}\right) \right] \right\} \quad (5)$$

Knowing the initial amount of component in the leaf, the mass transfer coefficients and the vapour pressure of the component, using Equation (5), the partial pressure that a component exerts at any time, t , during the SDE process can be determined.

The Gomez–Thodos vapour pressure estimation method was used to evaluate the value of the vapour pressure, P [11].

2.5. Heating-Up Period

Upon initial entry of steam into the plant bed, i.e., time zero, there would be an increase in the temperature of the bed from the ambient temperature that would subsequently increase with time to approach the steam temperature. Inclusion of this transient packed-bed heat transfer component, i.e., the heating-up period, into the proposed model, would better depict the SDE process than traditional models that do not compensate for this. Assuming no internal particle resistance to heat transfer, a solution presented by McGaw proves useful [12]:

$$\frac{\partial T_{\text{steam}}}{\partial y} = T_{\text{steam}} - T_p = \frac{\partial T_p}{\partial z} \quad (6)$$

where T_{steam} and T_p are the steam and bed temperatures relative to the initial bed temperature.

$$z = \frac{H_v}{C_p(1 - \epsilon)} \left[\theta - \frac{\epsilon s}{u} \right] \quad (7)$$

$$y = \frac{H_v s}{C_{\text{steam}} u} \quad (8)$$

H_v is the volumetric heat transfer coefficient, θ is the contact time interval, ε is the bed porosity, s is the depth of the plant bed, u is the superficial velocity of steam in the bed, and C_{steam} and C_p are the heat capacities of steam and the bed, respectively.

Then following McGaw, the following correlation was used to evaluate heat transfer coefficient [12]:

$$St = 0.72 Re^{0.3}, \quad (9)$$

Referring to Equations (6) to (9), the boundary conditions are:

$$\begin{aligned} \text{When } y = 0, T_{\text{steam}} &= T_{\text{steam},i} \\ z = 0, T_p &= 0 \end{aligned}$$

where $T_{\text{steam},i}$ is the inlet steam temperature.

Klinkenberg's proposed solution to these Equations in terms of error functions was said to be the most straightforward:

$$\frac{T_p}{T_{\text{steam},i}} = \frac{1}{2} \left[1 + \operatorname{erf} \left(\sqrt{z} - \sqrt{y} - \frac{1}{8\sqrt{z}} - \frac{1}{8\sqrt{y}} \right) \right] \quad (10)$$

This solution can then be integrated into the proposed SDE model by expressing the vapour pressure, P , as a function of time and temperature. In this way, the effect of the heating up period would be accounted for.

2.6. Application of the Model

Equations (1) to (10) relate to the basic heat and mass transfer processes that occur when steam is contacted with a single leaf over time. However, the processes of steam distillation involves the passage of steam through a vertical packed bed of plant material over a lengthy period.

Thus, at the beginning of the process, as the steam moves upwards through the plant bed, the simultaneous component transfer processes continue to take place, extracting specific amounts of components from successive layers. This process continues until the mixed vapour exits to the condenser where the water/oil mixture is condensed.

As more steam enters and contacts each layer in a similar manner and fixed amounts of oil components are removed from each layer, this process continues until all the oil constituents are extracted from the elements making up the plant charge.

Resultantly, concentrations of the components in the leaves and steam vary with the height of bed and contact time.

To model this, elemental analysis can be applied to take the basic equations and apply them throughout the height of the bed and the full time of extraction.

McGaw first employed this method of elemental analysis to predict the heat transfer in a cross-flow moving packed bed heat exchanger [12]. In adapting this approach to the proposed SDE model, the extraction bed is first divided into a finite number of $M \times N$ elements, based on dimensions of the extraction bed, the physical properties of the raw material, and the process flow conditions of the inlet steam.

M is the number of horizontal elements representing the height of the bed, each of which was estimated to comprise of one leaf.

N , the total number of vertical elements is effectively the extraction time dimension. Each vertical element/interval represents the residence time of the steam in the extraction chamber within the total extraction period.

The simulation starts from time zero by carrying out a set of M horizontal calculations up through the bed resulting with the oil content in the steam increasing as it passes through, and the reduced oil contents in each layer being recalculated as a result of the transfer.

New steam is passed through the bed for the second of the N vertical elements, and so on. Thus, for any (m, n) element, the following general equation applies

$$p(m, n) = p(m - 1, n) + P_{X_r} \left(1 - e^{\left(\frac{-kA_e t_{r,e}}{V_e} \right)} \right) \times \left(1 - \frac{8}{\pi^2} \left[\exp\left(-\frac{Dt_{r,e}\pi^2}{H^2} \right) + \frac{1}{9} \exp\left(-\frac{9Dt_{r,e}\pi^2}{H^2} \right) + \frac{1}{25} \exp\left(-\frac{25Dt_{r,e}\pi^2}{H^2} \right) \right] \right) \quad (11)$$

where, A_e : interfacial area of each element, m²

V_e : volume of steam surrounding each element, m³

m: horizontal layer within the extraction chamber

n: residence time of steam passing through the extraction chamber within the total extraction time

$t_{r,e}$: solvent residence time per element, s

H: Average leaf thickness

x_r , the fraction of oil that remains in the element, is defined according to the following equation:

$$x_r = \frac{8x_0}{\pi^2} \left[\exp\left(\frac{-D\pi^2 t_{r,e}}{H^2} \right) + \frac{1}{9} \exp\left(\frac{-9D\pi^2 t_{r,e}}{H^2} \right) + \frac{1}{25} \exp\left(\frac{-25D\pi^2 t_{r,e}}{H^2} \right) \right] \quad (12)$$

P is determined as a function of time and temperature by way of Equations (7) to (10).

3. Materials and Methods

To verify the model, we executed an experimental programme using *Ocimum basilicum* leaves grown in a tropical environment using the small scale pilot plant shown in Figure 1. The leaves were partially dried with ambient air to a moisture content of ~74% dry basis before being charged to the extraction chamber. The maturity stage and drying conditions for the basil used in the experimental work were chosen as a result of preliminary bench scale SDE experiments on the same source of basil used in work described in this paper and presented by Maharaj and McGaw [13]. After closing off the extraction chamber, steam was passed through the bed for 2 hours during which time samples of the essential oil from the material was collected from the oil separator every 10 minutes. Each run was for ~2 hours because the bulk of oil was collected in that time.

The chemical analysis of the basil oil produced in the experimental programme was carried out using Gas Chromatography-Mass Spectroscopy (GC-MS). There are many components present in basil oil, but this study limited the analysis to six components that comprised a mix of minor components - α -pinene, α -terpinene and geraniol -, and major components - eucalyptol, linalool, and eugenol. The six components were the similar to the components identified in the work of Hasegawa et al., Özcan and Chalchat, Sanda et al., Silva et al., Kéita et al. and Schulz et al. on *Ocimum basilicum* [14–20]. Since the named components were identified in multiple studies, this was used as the basis to qualify them as quality defining components comprising of a mix of major and minor components.

Percentage compositions, for six identified components, were determined using a standard mixture with variation coefficients being less than 5%.

4. Results

The extraction of the following basil oil components: α -pinene, α -terpinene, eucalyptol, linalool, geraniol, and eugenol were simulated from the model detailed in Section 4 using MATLAB. The external mass transfer coefficients, k, and the internal diffusion coefficients, D that modelled the removal of the components were determined using an optimized fit between the predicted extraction profile and the experimental results. The vapour pressures at the surface of the leaves were calculated using the Gomez–Thodos method, as described in Section 2.4. The k and D values are presented in Table 1 and will be discussed further in Section 5.1.

The extraction data for each oil component is shown in Figures 3–8, where it is seen that the predicted curves are generally quite close to the experimental data. For α -pinene and α -terpinene,

however, it can be seen in Figures 3 and 4 that while the model-predicted curves fitted the majority of experimental data points, it predicted a faster initial rate of extraction compared to the experimental data. α -Pinene and α -terpinene are minor components making up the oil and are highly volatile; yet, their transfer in the initial stages is inhibited. Potentially, there was preferential extraction of other components that were present in higher quantities namely, eucalyptol, linalool, and eugenol (Figures 5, 6 and 8). For these components, there was good agreement between the predicted values and the experimental data. In factoring the lack of fit for the minor components, It is also possible that the process conditions, such as steam pressure, temperature and flow rate, needed to be varied. This process is necessary to change the nature of extraction taking place for these components, and this is an area that requires additional investigations.

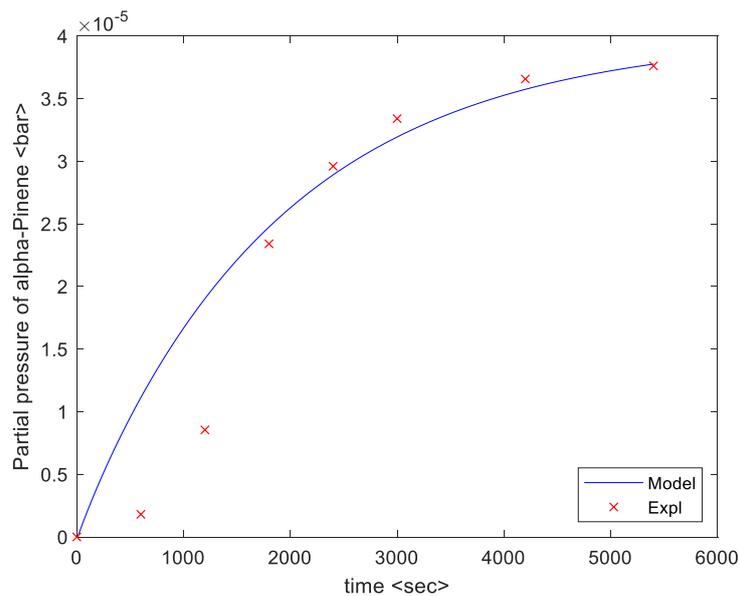


Figure 3. Modelling of α -Pinene.

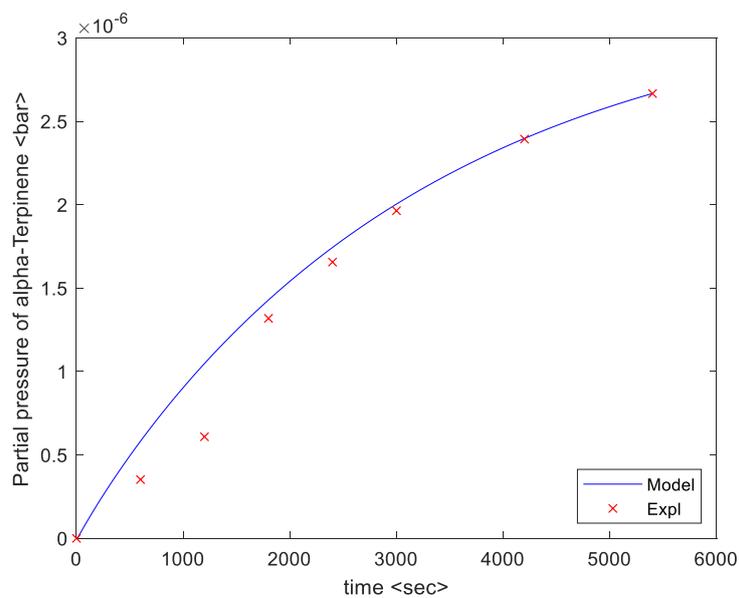


Figure 4. Modelling of α -terpinene.

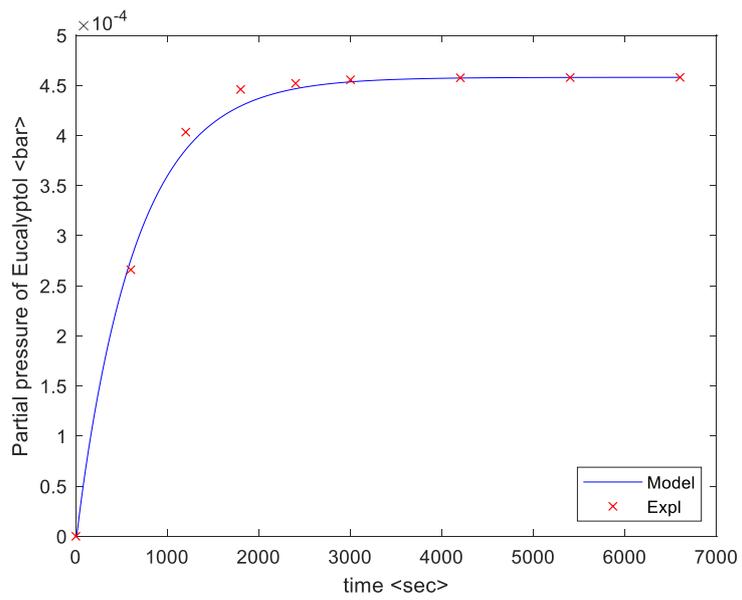


Figure 5. Modelling of Eucalyptol.

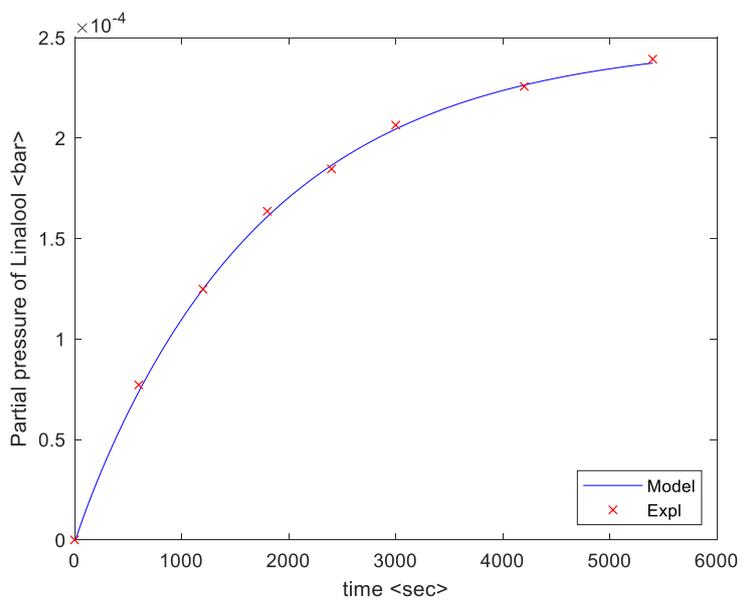


Figure 6. Modelling of Linalool.

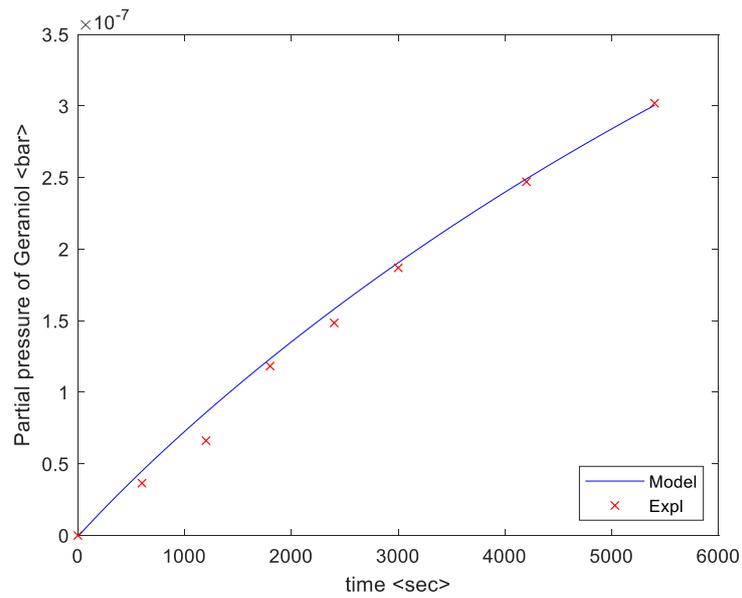


Figure 7. Modelling of Geraniol.

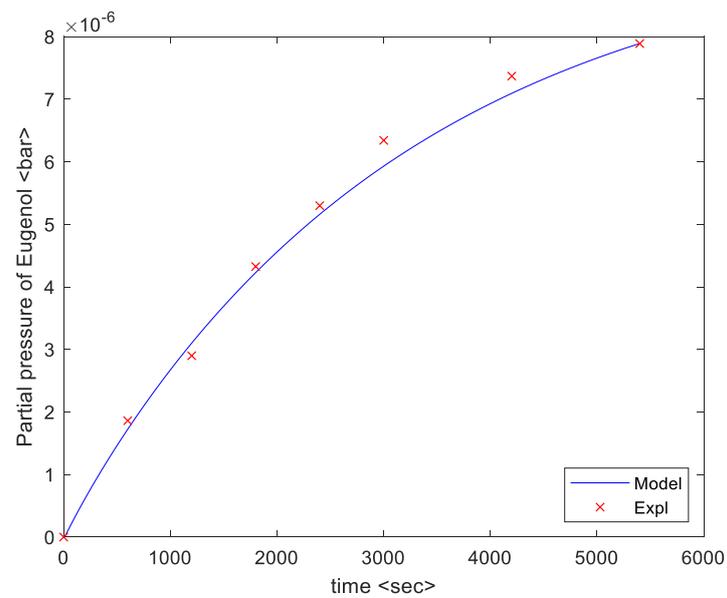


Figure 8. Modelling of Eugenol.

Table 1. Values of the predicted convective mass transfer (k) and diffusion coefficients (D).

Components	k Values* $10^{-5}/\text{ms}^{-1}$	D Values* $10^{-12}/\text{m}^2\text{s}^{-1}$
α -pinene	3.81	5.00
α -terpinene	2.59	2.98
Eucalyptol	5.14	6.99
Linalool	4.03	5.42
Geraniol	1.60	1.00
Eugenol	2.59	2.98

5. Discussion

5.1. Comparison of the Proposed Model with Existing Models

SDE is a traditional process for many developing countries that support the natural extracts industry. Mathematical models have been developed to allow for control and optimization of the process and are based on predicting the final quantum of the extract obtained. This assumes the oil to be a homogenous mixture and forms the premise of the proposed models which will now be considered.

Romdhane et al. developed a kinetic SDE model that treated all the oil constituents as one constituent solving for an overall mass transfer coefficient with units $\text{g oil h}^{-1}\text{m}^{-2}$. While there are no mass transfer coefficients to compare with the ones presented in this study, Romdhane et al. did recognize two mass transfer regimens that are common to the theory presented—surface extraction and diffusion to the surface [3].

The SDE model proposed by Cassel et al. was based on Fick's law in unsteady-state for a one-dimensional rectangular geometry and assumed the essential oil to be homogenous and constant, requiring only the diffusion coefficient to predict the experimental curve. For basil, the value of D was of the order of $10^{-11} \text{ m}^2\text{s}^{-1}$. While their value is higher than that reported in Table 1, this disparity arises out of the different assumption taken for this study where the oil is not considered to be homogenous [5,6].

Xavier et al. applied Sovova's broken and intact cell (BIC) model to SDE which also recognizes two extraction regimes, i.e., diffusion of oil to the surface and extraction of oil by the solvent, similar to this study [7,8]. However, this BIC model evaluates different parameters to this study, namely the solid and solute phase mass transfer coefficients, with each having units m^3s^{-1} . While the BIC model is very robust, application of the model follows very elaborate guidelines that involve developing three equations from eight possible mechanisms with up to seven experimentally determined parameters. One of the disadvantages of this model is that the parameters cannot be determined from a single experiment.

Compared to other models, the results presented in this study show that each component would have a relationship with the solvent. The proposed model applies a rigorous analysis based on fundamental engineering relationships with two adjustable parameters while incorporating a heating-up period which is inherent in any start-up SDE operation. It respects the two extraction regimes that have been proposed in the previous studies described and also mathematically link these phenomena to reflect their synchronous operations. However, the proposed model does not assume the oil to be a single component and allows for predicting the extraction of different oil constituents. As far as can be discovered, this simplification in treating the extract as a single pseudo component is common to all currently existing SDE models. There are no available comparative k and D values for each component presented in Table 1. Meanwhile, the results presented by Priyanka et al. show that the parameters are of the same order of magnitude for other extraction models that solve for external mass transfer and internal diffusion coefficients [21].

Considering that the internal diffusion to the surface involves a transfer through intra-cellular anatomy, it is likely that this is the major resistance compared to the vaporization and convective transfer from the surface. This process also accounts for the low magnitudes of D that were determined for each component. The prevailing mathematical models favour approaches that consider only the diffusion coefficient. However, the study conducted by Meziane et al. found that neglecting the essential oil composition in the vapour phase renders them unrepresentative especially if the SDE operation requires a change in the steam flow rate [9].

The characteristics of the extraction system used were built logically into the proposed model to mathematically model the extraction of the chemical components monitored during SDE. After accounting for these factors in the developmental stages, the model was found to result in reliable fits between the experimental and model-predicted data, giving values of both convective mass transfer (k) and diffusion coefficients (D).

Incorporating this model into the process, as well as the quality estimation techniques offers more advantages in predicting and controlling the process outcome. This is compared to the conventional methods of quality assessment by the final yields of the extract obtained. The proposed model provides a comprehensive framework of the SDE process and can be used to guide further research in this area.

The model was explicitly applied for leaves, thereby defining the geometry for internal diffusion. Its application for other particle shapes, such as for ground root crops, for example, ginger, could easily be adapted using the Crank equation for spherical shapes [10].

5.2. Recommendations

1. The effect of the steam pressure, temperature, and flow rate could be investigated by varying the process conditions to observe the nature of extraction of the components and verify our analysis further.
2. The proposed model was developed using Raoult's law which considers an ideal liquid solution that also follows the ideal-gas laws. Even though Treybal (1981) stated that for engineering purposes, this law could be applied, further model development should take this factor into account [22].

6. Conclusions

The proposed SDE model for the extraction of oil components appears to be accurate for the extraction system used. There was a good match between the model predicted values and the experimental data. The magnitudes of the predicted values of the convective mass transfer (k) and the diffusion (D) coefficients, ranged from 10^{-5} for k and $10^{-12} \text{ m}^2\text{s}^{-1}$ for D .

For some of the more volatile components such as α -pinene and α -terpinene, the model predicts a faster initial extraction rate than the experimental values. Potentially, the preferential extraction of light and volatile components present in larger quantities took place.

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Conflicts of Interest: The authors declare no conflict of interest.

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