The kinetic modelling of a steam distillation unit for the extraction of aniseed (*Pimpinella anisum*) essential oil

Mehrez Romdhane¹ and Chedly Tizaoui²

¹Unité de Recherche Réacteurs Chimiques et Commande des Procédés Industriels, Ecole Nationale d’Ingénieurs de Gabès, 6029 Gabès, Tunisia
²School of Engineering, Design & Technology, University of Bradford, Bradford, BD7 1DP, UK

Abstract: A simplified model that considered the oil constituents as one constituent, anethole, the major component, was used to describe the mass transfer of steam extraction of aniseed essential oil. The model can be used to optimise and control the process. Depending on the oil content, two mass transfer regimes were identified (i) the first one corresponds to an unsaturated surface extraction and (ii) the second corresponds to the slower transfer of oil from the deeper parts of the material to the surface, which may be due to concentration gradients and chemical bonding. The model was validated by experimental data obtained from a pilot-plant system. Solid-steam mass transfer coefficients were determined and a critical oil content was found to limit the two mass transfer regimes. The value for this critical oil concentration (*x₈*) was found to be 0.011 (g oil g⁻¹ solid). In addition, an optimal operating pressure of 200 kPa was found to give maximum extraction yield.

Keywords: aniseed; *Pimpinella anisum*; essential oil; steam distillation; extraction

NOTATION

- \(a\): Specific interfacial area per unit mass of bed (m² kg⁻¹)
- \(a₁, b₁\): Constants, see eqn (15)
- \(a₂, b₂\): Constants, see eqn (16)
- \(Gₜ\): Mass flow rate of steam (kg s⁻¹)
- \(K\): Overall mass transfer coefficient (g oil h⁻¹ m⁻²)
- \(mₗₘ\): Mass of the seeds (g)
- \(m_{ₒₒ}\): Mass of oil extracted by ethyl ether from the aqueous phase (g)
- \(m_{ₒₗ}\): Mass of oil contained in the seeds (g)
- \(m_{ₒₒₘ}\): Mass of the extracted organic phase (g)
- \(mₗ₟\): Total mass of oil extracted (organic and aqueous) (g)
- \(Mᵢ\): Mass of the inert part of the seeds (excluding the essential oil) (kg)
- \(N\): Rate of extraction (g oil h⁻¹ m⁻²)
- \(P\): Pressure (kPa)
- \(Pₒ\): Tension vapour of anethole (kPa)
- \(s\): In-column steam to material ratio (kg steam kg⁻¹ material)
- \(t\): Time (h)
- \(T\): Temperature (K)
- \(Vᵢ\): Column volume (m³)
- \(x\): Seeds oil fraction by mass (g essential oil g⁻¹ seeds)
- \(x₈\): Critical oil content by mass (g essential oil g⁻¹ seeds)
- \(y\): Steam oil fraction by mass (g essential oil g⁻¹ steam)
- \(Yᵢ\): Extraction yield
- \(α\): Proportionality parameter (kPa⁻¹)
- \(ε\): Column voidage
- \(ρₚ\): Steam density (kg/m³)
- \(Ω\): Surface area available for mass transfer (m²)

Subscripts

- 0: Initial
- f: Final
- in: Input
- T: Total

Superscript

- *: Equilibrium

1 INTRODUCTION

Essential oils are used in many industries such as the pharmaceutical and cosmetics industries. They are present in plants at low concentrations which would require extraction techniques of high performance in order to achieve high yields. Various techniques have been developed, including solvent extraction,¹⁻³ supercritical fluid extraction,⁴ hydrodistillation and steam distillation,¹³⁵ the use of superheated and
subcritical water\textsuperscript{6–9} and combinations of the previous techniques with others such as ultrasound and microwave-assisted processes.\textsuperscript{10,11}

None of these processes is universal; in effect each process presents particular advantages and disadvantages when used for a particular material. Nevertheless, hydrodistillation and steam extraction processes are the most used in industry and have been used since antiquity. They do not only produce high quality oil but they are simple to operate too. They can be used in either batch or semi-continuous configuration, they are environmentally friendly and are relatively safe to operate compared with other processes.

In the present study, a pilot plant was designed and constructed in order to test the steam distillation process for the extraction of aniseed essential oil. Optimal operating conditions were determined in order to achieve high extraction yields. A solid-steam mass transfer model was developed and mass transfer coefficients were determined as a function of steam pressure. Green seeds of commercial Tunisian aniseed were chosen as the solid matrix.

Aniseed (\textit{Pimpinella anisum}) is a plant cultivated throughout Europe and north Africa. It has been used since antiquity in medicine and cooking. It has a characteristic odour and a special sweet taste. It is listed in many official standards such as the British and the European pharmacopoeias. Aniseed promotes digestion, prevents flatulence and improves appetite. It has also antimicrobial and ovicidal activity.\textsuperscript{12,13} For medicinal purposes, aniseed is used as an ingredient in cough medicines and is reported to have diuretic and diaphoretic properties. It is used to treat dyspeptic complaints and catarrh of the respiratory tract. Aniseed contains about 2.4–3.5\% of essential oil.\textsuperscript{3,14} Anethole forms the chief constituent of the essential oil of aniseed, it accounts for 80–94\% of the total essential oil\textsuperscript{4,15,16} and is responsible for its taste and smell.

2 EXPERIMENTAL

The experimental unit is shown in Fig 1. It was designed and constructed at the National School of Engineering, Gabès, in collaboration with an industrialist from the region. The unit was mainly composed of a 50 dm\textsuperscript{3} stainless steel column (id = 266 mm) that contained five perforated plates (each perforation is of 2 mm diameter). The plates were placed on a stainless steel support. The maximum content of each plate was 2 kg of the solid material. Because the anised grains swell once in contact with steam and an expansion of the bed was inevitable, only 20–50\% of the total capacity of a plate was used. The column was insulated in order to minimise heat losses. Saturated steam was produced by an electrical boiler (18 kW max, 20 kg steam h\textsuperscript{−1} max and steam pressure 300–400 kPa). A pressure gauge was placed at the top of the column to measure steam pressure and a thermocouple was used to measure the temperature inside the column. Once the system reached steady state both temperature and pressure remained constant. The mixture of steam and essential oil at the column outlet was condensed in a glass condenser that was designed to cope with various operating conditions of steam flow rates and pressures. The condensate was collected in a sealed bottle and further cooled in a water and ice-cooling bath. Two liquid phases were obtained:

(1) An organic phase composed mainly of the extracted essential oil to which was added sodium sulfate to eliminate any traces of water.

(2) An aqueous phase (aromatic water) that contained an important amount of essential oil either dissolved in water or being dispersed in fine droplets (25\%). The essential oil found in the aqueous phase was extracted by ethyl ether and then distilled.

In all experiments carried out in this work, the amount of the aniseed was divided evenly between the five plates and the extraction yield was defined by:

\[
Y_E = \frac{m_{oo}}{m_{as}} = \frac{m_{oo} + m_{oa}}{m_{as}}
\]

where

\(Y_E\) extraction yield

\(m_{oo}\) total mass of oil extracted (organic and aqueous) (g)

\(m_{as}\) mass of the aniseed (g)

\(m_{oo}\) mass of the extracted organic phase (g)
3 RESULTS AND DISCUSSION
3.1 Overall and plate yields
The overall yield obtained was determined by measuring the total amount of oil extracted (in both organic and aqueous phases) after 2.5 h at a pressure fixed at 200 kPa and steam flow rate of 6 kg h$^{-1}$. The total amount of the aniseed was 2 kg (ie 400 g per plate). Two experiments were conducted at the same conditions and showed reproducible results (2.46% and 2.63%), giving an average yield of 2.55%. This extraction yield of the Tunisian aniseed used in this work falls in the same yield range of aniseed extractions found from other regions such as Europe, Russia and Egypt.$^{3,17–19}$

It is important to know how well the equipment is performing internally. Indeed poor hydrodynamics due to channelling can lead to poor and variable extraction performance between plates. This can therefore affect the mass transfer kinetics and the overall performance of the equipment. The aim of part of this work was to determine a method to measure the internal performance of the equipment. The method consisted of the measurement of the oil retention in each plate and to determine its variations between the five plates. At the end of an extraction experiment, where the five plates were used, each plate was re-extracted for about 2h separately at the same conditions of pressure and steam flow rate as in the initial extraction experiment. Results of two experiments ($P = 200$ kPa and $m_{\text{m}} = 2$kg) were reproducible and are presented in Table 1 and Fig 2. The oil retention is the percentage of oil obtained in the re-extraction experiment from each plate compared with the total amount of oil extracted (for example the oil retention in plate 1 in Experiment 1 is $100 \times 0.27/(49.21 + 5.40) = 0.49\%$).

Figure 2 shows the oil retention in each plate after the first extraction, indeed it mirrors the internal performance of the equipment. Since the oil retention in plate 3 is higher than that of plate 4, one can conclude that plate 3 has shown poor performance in the first extraction. This may be explained by the poor internal hydrodynamics of steam which resulted in lower steam/solid contact. Although the described functioning abnormality concerns only plate 3, the overall trend (except plate 3) is expected, as is explained in the following paragraph.

The average amount of oil retained by plate 1 is about 0.5% and by plate 5 is 3.9%. When extraction takes place, the fresh steam, in contact with the first layers (ie plate 1) of the solid, extracts oil rapidly and effectively due to a high concentration gradient. The steam then becomes more and more charged with oil. It is expected that steam would reach a saturation point that limits the mass transfer. In addition, plate 5 had retained nearly half the total oil retained by all the plates (ie 3.9% of oil compared with a total 10.0% retention).

The total oil obtained by extraction and re-extraction gave an average value of 2.8% for the yield; this value is just higher than that obtained by one extraction but it is still in the same range of yields reported in the literature for aniseed from other regions.$^{3,14}$

Finally, the performance of the equipment seems acceptable since 90% of the oil is extracted in about 2h 30min. Nevertheless, it seems possible to minimise the plates’ oil retention at the end of an extraction by better sealing of the plates to the body of the column and cross-feeding with fresh steam.

3.2 Extraction kinetics
Figure 3 shows the variation of the extraction yield with time at various steam pressures and loads. Figure 3(a) shows that, for a given duration of the extraction, both the extraction kinetics and the final yield obtained after 2h 30min are enhanced if the pressure increases. After an extraction time of about

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**Table 1.** Re-extraction results

<table>
<thead>
<tr>
<th></th>
<th>Experiment 1</th>
<th>Experiment 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>$m_{\text{in}}(g)$ 1st extraction (2.5h)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Plate</td>
<td>1</td>
<td>2</td>
</tr>
<tr>
<td></td>
<td>3</td>
<td>4</td>
</tr>
<tr>
<td>$m_{\text{in}}(g$ per plate) Re-extraction (2h)</td>
<td>0.27</td>
<td>0.30</td>
</tr>
<tr>
<td>$m_{\text{in}}(g$ Re-extraction</td>
<td>5.40</td>
<td>5.88</td>
</tr>
<tr>
<td>Oil retention (%)</td>
<td>0.49</td>
<td>0.55</td>
</tr>
</tbody>
</table>
2h 30min the yield increases from 1.8 to 2.1% for pressures of 140 and 200 kPa respectively. Figure 3(b) shows that for a given pressure (200 kPa), lower charges yield higher kinetics at earlier extraction times. But after longer periods of time, the extraction yield reaches a similar value (ie 2.1%) for the different loads. Since it was shown that the steam pressure and the column load affect the final yield and the extraction kinetics, it is important then to optimise the operating conditions of the system. A compromise needs to be considered between the number of extractions made in 24h, the yield of extraction and the amount of material used (and need to consider carefully the fluctuations in the price of the raw material and the quantity and quality of oil extracted).

It was found that the amount of oil extracted from the aqueous phase was considerable and in some cases has reached about 25% of the total oil extracted. Indeed, at 140 kPa and after 2h 30min, the amounts of oil extracted in the organic and the aqueous phases were 27.54 g and 8.88 g respectively. Therefore, it is essential to consider the conditions at which the amount of oil in the aqueous phase is kept minimal in order to avoid extra separation units.

### 3.3 Effect of pressure on the extraction yield

The effect of pressure on the extraction yield (2h 30 min duration) was studied. Figure 4 shows the results obtained for pressures between 140 and 250 kPa. This figure shows that the yield reaches a maximum at 200 kPa (2.5%) and then declines when pressure increases. For pressures less than 200 kPa the increase in the yield with pressure may be explained by the fact that an increase in the saturated steam pressure would result in an increase in temperature that increases the driving force for mass transfer as a result of an increase in the diffusion of solutes. Further increase in the pressure to more than 200 kPa results in a decrease in the yield that is most probably due to degradation of the oil components at higher temperatures.

### 4 QUALITATIVE STUDY

At 20°C the refractive index of the extracted oil was 1.4908. This value is in accordance with the European standards of pharmacology. Analysis of the extracted oil carried out by plate chromatography (Kiesel F 1500/LS254; ether/cyclohexane 1:4) at various times of the extraction showed that the oil composition changed slightly but a major component (anethole) dominated the overall composition. Although this method gave an indication about the evolution of the oil composition during operation, other analysis methods (HPLC, GC/MS etc) would give better and more accurate results for the oil composition, but this was not the objective of this research.

### 5 MODELLING

Only very few authors have attempted to model the kinetics of steam oil extraction processes. This may well be due to many factors related to both the difficulty of resolving the problem of mass transfer due to the complex nature of the system (internal and external porosity, solid dimensions, oil locality etc) and to the fact that the industry tends to use traditional methods of steam extraction and little importance is given to the control and optimisation of the process. Many firms rely for their steam production on traditional technologies. These technologies are not well controlled or optimised and are not environmentally friendly (ie not energy efficient). A fundamental understanding of the steam oil extraction processes is therefore required.
Denny\textsuperscript{20} has analysed the processes of diffusion of \textit{Lavandula angustifolia} and \textit{Mentha piperita} for extra-cellular oils and \textit{Eucalyptus polybractea} for intra-cellular oils. He proposed a methodology for each category to optimise the extraction time. Morin \textit{et al}\textsuperscript{21} have proposed first order kinetics to model the extraction and Benyoussef \textit{et al}\textsuperscript{22} have studied the extraction by steam of the essential oil of coriander and have proposed two models, one based on diffusion and the other based on first order kinetics and they found that the latter model described better their experimental results.

In this study, a model based on a linear driving force which considers the composition of the oil as being made up of only the major component, anethole (anethole makes up more than 90% of the oil), has been used to describe the aniseed/steam mass transfer process. The model and the experimental data were used to determine the mass transfer parameters including an overall mass transfer coefficient.

5.1 Theoretical study

5.1.1 Assumptions

The following assumptions were made:

(i) The essential oil is considered as only one component (ie anethole).
(ii) The pressure and temperature are constant along the column.
(iii) The aniseed grains are considered to behave like a porous material.
(iv) The mass transfer is similar to that between a porous material and a gas.
(v) The bed is considered fixed and no movement of the particles due to the flow of steam is considered.
(vi) The particles are spherical and do not change either their shape or their size during the extraction process (however at the end of an experiment an increase in seeds’ volume was observed).
(vii) Initially, the composition in oil is homogeneous.
(viii) The internal condensation of steam is negligible.
(ix) The diffusion of steam inside the grains is negligible.
(x) The assumption of a well-mixed reactor is applied to the current system.

5.1.2 Mass balance

The overall oil mass balance made over the column boundaries gives:

\begin{equation}
G_{ol}y_{in} - G_{ol}y(t) = \frac{dm_{ol}}{dt}
\end{equation}

where \(m_{ol}\) is the mass of oil contained in the column, \(y_{in}\) is the input essential oil fraction by weight, \(y(t)\) is the steam essential oil fraction by mass in the outlet of the column at a time \(t\) and \(G_{s}\) is the mass flow rate of steam (kg s\(^{-1}\)).

The steam feeding the column is fresh and does not contain the essential oil, therefore \(y_{in} = 0\) and eqn (2) changes to:

\begin{equation}
y(t) = -\frac{1}{G_{s}} \frac{dm_{ol}}{dt}
\end{equation}

Consider that \(M_{i}\) is the mass of the inert part of the seeds (excluding the essential oil) and \(m_{as}\) is the mass of the seeds. At any time \(t\) the relationship between \(M_{i}\) and \(m_{as}\) is given by:

\begin{equation}
M_{i} = m_{as}(1 - x(t))
\end{equation}

where \(x(t)\) is the fraction by mass of oil in the seeds at a time \(t\).

The mass of oil in the column at a time \(t\) is then given by:

\begin{equation}
m_{ol} = m_{as}x(t) + \varepsilon \rho_{s} V_{C} y(t)
\end{equation}

\begin{equation}
= M_{i} \left\{ \frac{x(t)}{1-x(t)} + \varepsilon \rho_{s} V_{C} y(t) \right\}
\end{equation}

where \(\varepsilon\) is column voidage, \(\rho_{s}\) is steam density and \(V_{C}\) is column volume. the relationship between \(x(t)\) and \(y(t)\) is then:

\begin{equation}
dm_{ol} = M_{i} \frac{dx}{(1-x)^2} + \varepsilon \rho_{s} V_{C} dy
\end{equation}

The mass balance is rewritten as:

\begin{equation}
-\frac{G_{s} y dt}{(1-x)} = \frac{dx}{(1-x)^2} + \varepsilon \rho_{s} V_{C} dy
\end{equation}

Integration of eqn (7) between time 0 and a time \(t\) gives the value of the mass fraction \(x\) at time \(t\) as function of the steam oil fraction, \(y\). The initial fraction at time 0 is equal to the value of the maximum fraction of oil contained in the seeds (ie \(x_{0} = 0.028\)). Knowing that \(M_{i} = m_{as0}(1 - x_{0})\); \(m_{as0}\) is the initial mass of the seeds and \(x_{0}\) is the initial fraction of oil in the seeds. The following equation gives \(x(t)\):

\begin{equation}
x(t) = 1 - \frac{1 - x_{0}}{1 - \varepsilon \rho_{s} V_{C} \int_{0}^{t} y(t) dt + \frac{\varepsilon \rho_{s} V_{C}}{m_{as0}} (y_{0} - y)}
\end{equation}

where \(y_{0}\) is the outlet steam initial oil fraction, determined from experimental data.

5.1.3 Rate of extraction

The rate at which oil is transferred to steam can be described in terms of an overall mass transfer coefficient \(K\) and the difference between an average composition of the seeds, \(x_{s}\), and an equilibrium composition, \(x^{*}\), eqn (9).

\begin{equation}
\frac{dm_{ol}}{dt} = K \Omega (x - x^{*})
\end{equation}

where \(K\) is an overall mass transfer coefficient (g oil h\(^{-1}\)m\(^{-2}\)), \(\Omega\) is the surface area available for mass transfer (m\(^2\)) and \(x^{*}\) is an oil fraction in the seeds.
assumed in equilibrium with steam. It is difficult to determine \( x' \) directly, therefore it is proposed to assume that \( x' \) is proportional to the oil vapour pressure at a given temperature. The essential oil of aniseed contains mainly anethole (\( \geq 90\% \)), therefore the oil vapour pressure is assumed to equal that of anethole and \( x' = \alpha P^0 \) where \( \alpha \) is proportionality parameter, and \( P^0 \) is the pure vapour pressure of anethole at a temperature \( T \).

Equations (6) and (9) are combined to give the following differential equation:

\[
M\frac{1}{(1-x)^2} \frac{dx}{dt} + \varepsilon_\rho V_C \frac{dy}{dt} = -K\Omega(x - \alpha P^0) \tag{10}
\]

Consider specific mass transfer area, \( a \) (m\(^2\) kg\(^{-1}\)) defined by \( a = \Omega/m_\text{as} \) and an in-column steam to material ratio, \( \varepsilon \) defined by \( \varepsilon = \varepsilon_\rho V_C/m_\text{as} \). Equation (10) can be rewritten simply as follows:

\[
\frac{1-x_0}{(1-x)^2} \frac{dx}{dt} + \varepsilon \frac{dy}{dt} = -Kax + \alpha KaP^0 \tag{11}
\]

Note that eqn (11) can be further simplified if one considers that the oil content \( x \) is small compared with 1 (\( x_0 \) is the maximum value for \( x \) which is less than 0.03), eqn (11) is re-written in the following simpler form:

\[
\frac{dx}{dt} + \frac{dy}{dt} = -Kax + \alpha KaP^0 \tag{12}
\]

Values for \( Ka \) and \( \alpha \) were determined by fitting the experimental results to eqn (11). Indeed, graphical representation of the left-hand side of eqn (11) as function of \( x \) would result in a line of slope \(-Ka\) and intercept \( \alpha KaP^0\).

\( P^0 \) is the pure vapour pressure of anethole, the dominant component (>90%), at a given temperature \( T \). The variation of \( P^0 \) with temperature follows an exponential form, \( P^0(\text{kPa}) = 40\,433\,539.6 \exp(-6545.7/T) \) where \( T \) is the temperature in Kelvin.\(^{23} \)

### 5.1.4 Determination of the mass transfer coefficient and the proportionality parameter \( \alpha \)

It was possible to determine values for the group \( Ka \) using experimental data coupled with the model. From this group, the mass transfer coefficient \( K \) is determined if the specific area, \( a_s \), is known. The aniseed particles were assumed to be spherical and their average diameter was determined using the sieving method; a value of 3mm was obtained. The particle density was also determined and was found to be 273 kg m\(^{-3}\). The specific mass transfer area, \( a_s \), is then calculated to be 7.32 m\(^2\) kg\(^{-1}\). The overall mass transfer coefficient, \( K \), was determined for steam pressures of 140 and 200 kPa, which correspond to temperatures of 109 \(^\circ\)C and 120 \(^\circ\)C respectively. Once values for \( K \) were determined, values for the parameter \( \alpha \) were also determined.

### 5.2 Results and discussion

#### 5.2.1 Rate of extraction

From data obtained from extraction experiments, a curve of the oil content in the seeds as function of time was plotted. The data were converted to rates of extraction, expressed as \( N \) (kg oil extracted h\(^{-1}\) m\(^{-2}\)), and plotted against seeds oil content, as shown in Fig 5. This was done by measuring small changes in oil content, \( \Delta x \), for corresponding small changes in time, \( \Delta t \), and calculating the rate as \( N = -M_i \Delta x/\Omega \Delta t \). Figure 5 shows, for two pressures, two distinctive parts of the rate-of-extraction curve. The two parts are linear with two different slopes. The change in slopes is partly due to changes in the mechanisms of mass transfer and partly due to changes in the surface area from which oil evaporates. It is assumed that part AB of the curve represents easily accessible oil (extra-cellular surface oil) and part BC represents a rather difficult oil to access (intra-cellular deeper inside the particle or chemically bonded oil). The surface oil becomes scarce as evaporation proceeds and oil-free spots may appear upon the surface and occupy increasingly larger proportions of the exposed surface as the extraction proceeds. This leads to smaller area available for transfer and gives rise to the first part of the rate-of-extraction curve (AB); this period of the extraction process corresponds to unsaturated surface extraction. The unsaturated surface extraction region is bounded by a critical oil content \( x_B \), which bound the lower limit of this region. This critical concentration appears to not depend on pressure since the same value is obtained for the two pressures considered in this study (\( x_B = 0.011 \)) and maybe is characteristic of the material, Fig 5. On further extraction, the rate at which oil may move through the seeds, as a result of concentration gradients that exist between the deeper parts and the surface, is the controlling step. As the oil concentration is lowered by the extraction process, the rate of internal movement of the oil decreases. It may be that oil evaporation takes place beneath the surface of the solid, due to temperature and concentration gradients (ie at 120 \(^\circ\)C, \( P^0\text{anethole} = 2.33 \text{kPa} \)), in a zone which retreats deeper into the solid as the extraction process proceeds. In consequence, the rate of extraction falls more rapidly than before, as from B to C (Fig 5). The point C is reached after 2h 30min. However, if the experiment were carried out for a longer time, the point C should be obtained at the intersection of the prolonged line (BC) and the x-axis at which the rate of extraction is zero. The same trend of the rate-of-extraction curve is obtained for both pressures.

#### 5.2.2 Mass transfer coefficient and proportionality parameter

The mass transfer coefficient, \( K \), at different operating conditions was determined using the model developed earlier (eqn (11)) combined with experimental data. Values for \( K \) and \( \alpha \) are shown in Table 2. It was found that both the operating pressure and the load
have affected the mass transfer coefficient and the proportionality parameter, \( \alpha \). For a fixed load, \( m_{as} \), it was found that the mass transfer has increased by almost seven times when the pressure has changed from 140 to 200 kPa. This is explained by the fact that an increase in steam pressure results in a temperature increase, which facilitates oil diffusion from the inside of the seeds to steam by changing the physico-chemical characteristics of the oil such as viscosity, chemical bonding, etc. In addition it was experimentally observed that the structure of the seeds was modified following an increase in pressure. Indeed, larger channel openings were obtained following an increase in temperature, which made it easier for the oil to be transferred to steam and therefore enhanced the mass transfer. In contrast, for a fixed pressure, an increase in the load reduces the mass transfer coefficient by almost ten-fold when the load is changed from 2 to 5 kg. This can be explained by the reduction in the in-column steam to material ratio, \( \beta \). It was also found that the proportionality parameter \( \alpha \) increases slightly with pressure but decreases largely with the load.

5.2.3 Extraction time

The extraction time is an important parameter in the design of equipment; it determines the size of equipment needed for a given capacity. It is desirable to determine the time required to extract oil from larger batches of the material under the same extraction conditions used to conduct the experiments once the rate-of-extraction curve, such as in Fig 5, has been completely determined. In order to determine this time for such a curve, it is possible to proceed as follows:

By definition, \( N = -\frac{M_i}{\Omega} \frac{dx}{dt} \) (13)

Integration of eqn (13) between \( x_0 \) and \( x_f \), the initial and final oil contents, respectively gives:

\[
T = -\frac{M_i}{\Omega} \int_{x_0}^{x_f} \frac{dx}{N}
\]

where \( T \) is the total extraction time. Equation (14) is integrated analytically using the equations obtained from the rate-of-extraction results (Fig 5). \( N_i \) is linear in both regions, ie:

\[
N = a_1x + b_1 \quad \text{for } x \in [x_B, x_0] \quad (15)
\]

and

\[
N = a_2x + b_2 \quad \text{for } x \in [0, x_B] \quad (16)
\]

Table 3 summarises the various possible equations for \( T \) depending on the value of the critical oil content \( x_B \) with respect to \([x_0, x_f]\).

6 CONCLUSIONS

The configuration of the system used in the present work was effective to extract the essential oil of aniseed. In effect 90% of the oil was extracted in about 2h 30min compared with other techniques that require longer times, about 6h.\(^3\) It was found that pressure affects the yield of extraction and an optimum pressure of 200 kPa was found to achieve the highest yield. Analysis of the rate-of-extraction curves has revealed two regimes for mass transfer (i) the first one corresponds to an unsaturated surface extraction and is bounded by a critical oil content, \( x_B \), and (ii) the second regime corresponds to slower oil transfer from the deeper parts of the material to the surface. It was found that \( x_B \) has an experimental value of 0.011 g oil g\(^{-1}\) aniseed, which was independent of the pressure in the range 140–200 kPa. A theoretical model was proposed and was found to describe well the experimental data. The model and the experimental data were used to determine the mass transfer coefficient at different operating conditions. It was found that the mass transfer coefficient has increased by almost seven-fold when the pressure has changed from 140 to 200 kPa and has decreased when

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**Table 2. Mass transfer results**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Pressure (kPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>140</td>
</tr>
<tr>
<td>( m_{as} ) (kg)</td>
<td>2.0</td>
</tr>
<tr>
<td>( T ) (°C)</td>
<td>109</td>
</tr>
<tr>
<td>( P_0 ) (kPa)</td>
<td>0.015</td>
</tr>
<tr>
<td>( K ) (g m(^{-2}) s(^{-1}))</td>
<td>( 4.5 \times 10^{-4} )</td>
</tr>
<tr>
<td>( \alpha ) (kg m(^{-1}) K(^{-1}) s(^{-1}))</td>
<td>0.00216</td>
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**Table 3. Extraction time equations**

<table>
<thead>
<tr>
<th>Case</th>
<th>( t_f )</th>
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<tbody>
<tr>
<td>( x_0 &gt; x_i \geq x_B )</td>
<td>( t_f = \frac{M_i}{\Omega} \ln \left( \frac{a_1 x_0 + b_1}{a_1 x_i + b_1} \right) )</td>
</tr>
<tr>
<td>( x_0 &gt; x_B &gt; x_i )</td>
<td>( t_f = \frac{M_i}{\Omega} \ln \left( \frac{a_1 x_0 + b_1}{a_1 x_B + b_1} \right) + \frac{1}{\frac{\partial \Omega}{\partial x}} \times \ln \left( \frac{a_2 x_B + b_2}{a_2 x_i + b_2} \right) )</td>
</tr>
<tr>
<td>( x_B \geq x_0 )</td>
<td>( t_f = \frac{M_i}{\Omega} \ln \left( \frac{a_1 x_0 + b_1}{a_2 x_0 + b_2} \right) )</td>
</tr>
</tbody>
</table>

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the load is increased. Equations that determine the extraction time were also developed. These equations are of great interest in the design of larger systems operating at similar conditions to which this work has been carried out.

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