Research Article

A Novel Hybrid Method Based on Three-Stage Extraction and DEA-CCR Models for Selecting the Optimal Conditions for Citronella Oil Extraction

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Abstract
In the citronella oil extraction process by steam distillation, inefficient use of steam is the main cause of excessive energy consumption that affects energy cost and oil yield. This research is aimed to reduce the energy cost and increase the oil yield by studying the steam used in the process. The proposed method is the three-stage extraction model combined with the Data Envelopment Analysis developed by Charnes, Cooper, and Rhodes (DEA-CCR model). Although the three-stage extraction model has been widely used, there is no research integrating this model with the DEA-CCR model. It is well known that the DEA-CCR model is an effective tool to evaluate the efficiency of decision-making units/alternatives. The advantages of this research were presented as the calculation of the optimum distillation conditions, including the steam flow rate and the distillation time, were achieved as discussed in this article. The study was comprised of 3 parts. Firstly, the three-stage extraction model for citronella oil was formulated. Secondly, the results of the proposed model were calculated under different conditions, classified by steam flow rates from 5,000 to 60,000 cm^3/min for the distillation period of 15–180 min. Finally, the DEA-CCR model was utilized to evaluate and rank alternatives. The results expressed that the best condition for producing citronella oil was at the steam flow rate of 40,000 cm^3/min and the distillation time of 60 min. The optimal energy cost and percentage of oil yield were equal to 0.440 kWh/mL and 0.7%, respectively. When comparing to the experimental results, the percentage error of optimal energy cost and oil yield were slightly different, with a value of 0.98% and 0.85%, respectively. Moreover, the energy consumption was also reduced by 34.6% compared to the traditional operating conditions.

Keywords: Citronella oil, Three-stage extraction model, Steam distillation, DEA-CCR model

1 Introduction

Citronella oil is extracted from citronella grass, scientifically known as *Cymbopogon nardus*, which is native to Sri Lanka and India. The color of the oil ranges from clear to light yellow. Its benefits are for deodorizing, improving air purification, and killing germs. In Thailand, it is commonly used as an herb to repel mosquitoes and insects. All the citronella oil from steam distillation is pale yellow and has a mild odor [1]. The chemical constituents of citronella oil are Limonene citronellal, citronellyl acetate, neral, geranial, geranyl acetate, citronellol, and geraniol. These constituents are like the citronella oil from *C. Winterianus*. The citronella oil from *C. nardus* contains geranial in the chemical constituents but does not contain limonene [1]. In addition, citronella oil has a relative density of 0.8–0.9 [2] and a boiling point of

170 °C [3], [4].

In recently, there are many studies comparing the extraction of citronella oil by different methods. Danh *et al.* [5], [6] found that supercritical extraction technology. It is a promising technology to extract the components in a cleaner way, which higher purity than other technologies. Nevertheless, this technology uses high energy to compress and decompress supercritical systems. Compared to conventional solvent extraction technology, yields are high due to the selection of solvents and the interesting compounds to be extracted. However, hazardous solvents are sometimes required to ensure proper extract. This may cause environmental problems and the quality of the extracted oil can be affected depending on the selected organic solvent [7]–[9]. Furthermore, the oldest method, water distillation technology, is used to extract essential oils with high yield and high purity of the extract. Moncada *et al.* conducted a case study of distillation of lemongrass oil and citronella oil, comparing three distillation technologies: supercritical liquid, solvent distillation, and water distillation [10]. The results presented that water distillation had the lowest production cost and lowest environmental impact, with the lowest carbon emissions for extraction by distillation with full energy integration. Muttalib *et al.* [11] also found that the choice of solvent influenced the result of product extract in the extraction process by distillation to produce a good yield of lemongrass oil extract. Water was the best solvent extract to lower the toxic effect.

In industry, the extraction of essential oils by steam distillation is the most used method, because of its low installation costs, as well as low operating and maintenance costs [12]. In addition, other methods, such as the solvent extraction method may cause residues of solvents and non-volatile components. If volatility is subsequently treated, it adds additional costs and environmental risks. In steam distillation, essential oils are extracted at a temperature below the boiling point of this oil (170 °C) and the heat-sensitive compounds are separated from the plant. This process can produce a good quality of essential oils. The operation process is simple, safe, and environmentally friendly. Steam distillation also has the added benefit of protecting the volatiles from oxidation by replacing oxygen with water vapor while the volatile components are being condensed. Nevertheless, the disadvantage of this process is a high level of energy consumption. Therefore, the selection of steam distillation parameters, such as the steam flow rate and the distillation time is important, because, under the optimal conditions, the energy consumption of this process can be reduced.

Initially, our research was inspired by the production of citronella oil at a small plant in Yang Talat District, Kalasin Province. The plant had a capacity of 0.62 L of citronella oil per day, with a power consumption of 416 kWh. It was illustrated that this traditional distillation process consumed a lot of costs. The researchers then started by figuring out how to reduce costs by improving the production process. To improve the process, some methods were able to be used, such as re-distilling wastewater to recover the dissolved oil components [13]. It was resulted in increasing oil yield but the utility costs i.e., heating or energy costs, were also increased. Another method assumed as being worked was the use of the microwave technique to assist the distillation system [14]–[18]. This method could also improve the process, but it required additional equipment and construction funding. The research team then came up with ideas for improving the production process by reducing energy costs from the inefficient use of steam.

The optimization of energy consumption method is a method that uses energy efficiency as an indicator to evaluate energy consumption per essential oil yield. However, recent studies are demonstrated that this method cannot present a correlation between the rate of steam consumption, distillation time, and the essential oil yield [19]. Some studies used the steam distillation method for different types of oil and successfully demonstrated the optimum oil yield. Golmohammadi [20] optimized the production of citrus peel oil yield by an experimental method. Galadina [21] and Rezzoug [22] quantified optimal essential oils using experimental design and the surface response method. Kaya quantified the optimal *Myrtus cornunis* oil yield by designing an experimental method and the Taguchi technique [23]. Unfortunately, the main disadvantage of these methods was the necessity to perform multiple trials to find a reliable break-even point from the results of the studies. This repetitive process brought a waste of labor, energy, and capital. Thus, the research team was interested in finding a computational method that able to be used to predict the break-even point of production without the need for repeated experiments.

Several studies demonstrate the kinetics of the extraction of essential oils by steam distillation. Romdhane and Tizaoui [24] described two parts of the mass transfer. The first part involved the extraction of unsaturated surface areas, and the second part involved the slow mass transfer of the oil from the inside to the surface of the plant. Xavier *et al.* [25] described two types of cell classification: broken cells and completed cells. The oil contained in the broken cells was rapidly extracted, while that in the completed cells slowly spread to the plant surface. Milojević *et al.* [26] explained the principle of the essential oil extraction occurring in two mechanisms simultaneously: a) "washing" of essential oils on the external surface of plant particles, and b) the diffusion of essential oils from the inside to the external surface of plant particles. Cassel and Vargas [27] explained the principle of particle diffusion and mathematical modeling of the steam distillation process using Fick's law based on unstable conditions for one-dimensional rectangular geometry. Cerpa *et al.* [28] described the three-stage extraction model by classifying the extraction process into three stages, including i) thermal exudation of oil from glandular trichomes, ii) vapor and liquid of oil component equilibrium at the interface, and iii) the vapor phase oil mass transfer and oil condensation. In the present, there is a study of extracting an essential oil using this model compared to other models [27] and other research studies have worked to develop methods to control the quantity of oil extracted [29].

The difference in the three-stage extraction model compared to other models was that the entire extraction process was able to be presented mathematically. In this model, the oil mass flow rate of each stage could be calculated, including the oil mass flow rate of exudation in trichomes, the oil vapor mass flow rate at the interface and the oil condensation rate. This research used the three-stage extraction model to analyze the citronella oil extraction process and to calculate the variables under different conditions. The three-stage extraction model was advantageous as it was able to predict the extracted oil yield in different conditions where the steam flow rate and the distillation time were the main parameters of this calculation. In addition, citronella was a medicinal plant, and it had some physical properties such as boiling point and relative density suitable for extraction by steam distillation [1]–[3]. The three-stage extraction model was also a model derived from the oil extraction process by steam distillation. Therefore, this model was used to predict the results of the extraction of citronella oil in this research.

The Data Envelopment Analysis approach, developed by Charnes, Cooper, and Rhodes [30], (DEA-CCR), is a mathematical model for measuring the efficiency scores of decision-making units (DMUs) with multiple input and output variables [31], [32]. The efficiency scores of DMUs are calculated by maximizing the ratio of the sum of weighted outputs to the sum of weighted inputs. The maximum ratio of each DMU, which is not greater than 1, is defined as the efficiency score [31], [33], [34]. A DMU can be defined as being efficient if its efficiency score is equal to 1. In addition, if there is only one efficient DMU, no other ranking method is needed. Hence, DEA-CCR is a technique for evaluating the optimal value of alternatives. This is an interesting technique for evaluating the optimum conditions of citronella oil extraction calculated from the three-stage extraction model.

Combining this three-stage extraction model with DEA-CCR analysis is named the hybrid method, which has never been achieved before. Therefore, the objective of this study is to demonstrate this novel hybrid method based on a three-stage extraction model and the DEA-CCR model for determining the optimal steam flow rate and distillation time.

2 Background

2.1 *Schematic diagram of the extraction process*

The three-stage extraction process [11] can be illustrated by the flow diagram in Figure 1.

From Figure 1, oil transport from the plant to the vapor phase is considered to take place in three stages.

In stage 1, oil is initially inside glandular trichomes. In the thermal exudation stage, oil is heated by steam and transferred to the water-oil layer at the interface.

In stage 2, oil and condensed water form a water-oil layer wetting the plant. Then, oil components evaporate at the vapor-oil interface according to phase equilibrium.

In stage 3, evaporated oil components are transferred from the vapor-oil interface to the steam stream and condense.

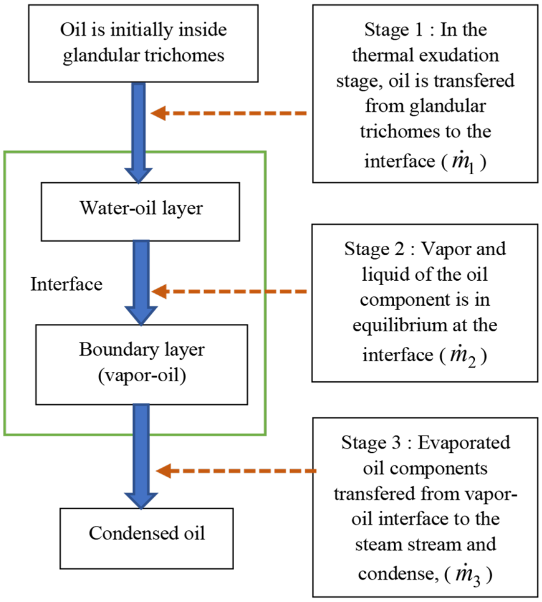

Figure 1: The schematic diagram of the extraction process.

2.2 *Modeling of essential oil extraction by steam distillation*

The model is applied for the extraction of essential oil with the following assumptions [28]: a) The system must attain constant temperature and pressure; b) All parts of the plant must be considered in a batch; c) The batch porosity must be constant; d) The vapor phase is perfectly mixed with a constant flow rate and the accumulation of oil in the vapor phase is negligible; e) All oils in the trichomes will be extracted in the process; f) Condensed water and essential oil are completely immiscible and g) The steam stream fed to the distillation vessel must be oil-free. These assumptions are defined to simplify the equations of the essential oil extraction process, which is classified into three stages as shown in Figure 1. The mathematical model was further determined in the next section.

2.2.1 Thermal exudation of oil from glandular trichomes

At first, the oil is still inside the glandular trichomes and is heated from the steam flow causing the thermal exudation stage. The mass flow rate of oil exudation is calculated as in the equation [28]:

$$\dot{m}_1 = -W\dot{G} = K_{tr} m_1 W \qquad (1)$$

where G = the oil mass inside the trichomes per mass of fresh plant in g/g, W = the fresh plant mass in g, and K_{tr} = the exudation kinetic constant in min^{-1}.

2.2.2 Vapor and liquid of the oil component equilibrium at the interface

Oil is released from the trichomes and evaporated at the vapor-oil interface along with the steam flow. The oil concentration in the vapor phase is given by the following equation [28]:

$$C^* = \frac{P^{0,os} M_W}{RT} \qquad (2)$$

where T = the temperature at the vapor-oil interface in °C, R = the universal gas constant and $P^{0,os}$ = the vapor pressure of the oil in the two-phase aqueous layer in kPa (obtained from Raoult's law, which is the sum of the product of mole fraction and vapor pressure of oil components, which are based on experimental results as shown in Table 1) [35]. The equation is

$$P^{0,os} = \sum_{i=1}^{6} x_i^{os} P_i^0 \qquad (3)$$

where x_i^{os} = liquid mole fraction of the essential oil component in the two-phase aqueous layer, P_i^0 = the vapor pressure of the essential oil components in kPa (obtained from Antoine's Equations) [36], M_W = the molecular weight of oil in g/mol. It can be expressed as in the following equation.

$$M_W = \frac{1}{P}\sum_{i=1}^{N} x_i^{os} P_i^0 M_{wi} \qquad (4)$$

where N = the number of oil components, M_W = the molecular weight of oil components in g/mol, which is shown in Table 1, P = the pressure of the oil in kPa, which can be obtained from Dalton's law, $P = P_W^0 + P^{0,os}$, P_W^0 = the vapor pressure of water in kPa.

The mass transfer rate from the vapor-oil at the vapor-oil interface must be along with the steam flow. The relation is presented as in the following equation [29]

$$\dot{m}_2 = K_{tr} m_1 W - \frac{K_g C^* m_2}{h\rho_{eo}}\left(1-\left(\frac{K_g m_2}{uh\rho_{eo}+K_g m_2}\right)\right) \qquad (5)$$

where h = the oil spots average thickness in cm, ρ_{eo} =

the essential oil liquid density in g/cm^3, K_g = the mass transfer coefficient in cm/min.

Rexwinkel [37] described the mass transfer of flow through inert beds, which could be expressed as, $Sh_p = 0.3Pe_p^{1.3}$, where Sh = Sherwood's number, and Pe = Peclet's number. In the case of $Re < 10$ and $Pe > 100$, the equation for mass transfer coefficient is given as follows:

$$K_g = 0.3\frac{D}{L}\left(\frac{LU}{D}\right)^{1.3} \tag{6}$$

where L = the characteristic length in cm, U = the steam velocity in cm/min which can be found from the equation of flow through a porous media as $U = \frac{Q}{A\phi}$, A = the cross-sectional area in cm^2, ϕ = the porosity, Q = the volume flow rate in cm^3/min, D = the gas diffusivity of binary air-hydrocarbon or non-hydrocarbon gas mixture at low pressures obtained from the Fuller-Schettler-Giddings equation [38]:

$$D_{AB} = \frac{10^{-3} T^{1.75}\left(M_A^{-1} + M_B^{-1}\right)^{1/2}}{P\left(\left(\Sigma v\right)_A^{1/3} + \left(\Sigma v\right)_B^{1/3}\right)^2} \tag{7}$$

where D_{AB} = the binary gas-phase diffusivity of A in B in cm^2/s, T_{abs} = the absolute temperature in Kelvin, P_{abs} = the absolute pressure in atmospheres in kPa, M_A, M_B = the molecular weight of A and B, respectively in g/mol, $(\Sigma v)_A$, $(\Sigma v)_B$ = the molecular volume in cm^3/g·mol

2.2.3 Vapor phase oil mass transfer and oil condensation

The vapor phase oil mass transfer and mass balance of oil after condensation must be considered according to the following equation:

$$\dot{m}_3 = \frac{K_g u C^* m_2}{u h \rho_{eo} + K_g m_2} \tag{8}$$

2.3 *Citronella oil compounds and their properties*

Nitangsam [1] described the citronella oil compounds and their properties, which were obtained by the experimental method and illustrated in Table 1. The main substances in citronella oil include citronellal, citronellyl acetate, geranial, geranyl acetate, citronellol, and geraniol. A composition ratio, molecular weight and vapor pressure as shown in Table 1 were used for the calculation of the three-stage extraction model in Equations (2)–(4).

2.4 *Data Envelopment Analysis (DEA)*

Let there be a set of n DMUs, where DMUj (j = 1, 2, 3, . . ., n) uses m different inputs to produce s different outputs, which can be denoted as x_{ij} = (1, 2, 3, . . ., m) and y_{rj} = (1, 2, 3, . . ., s), respectively. μ_{rd} and ω_{id} are the weights of outputs and the weights of inputs, respectively. For any evaluated DMU$_d$ ($1 \le d \le n$), the efficiency score E_{dd} can be calculated by the CCR model as in the following equation:

$$Max \sum_{r=1}^{s} \mu_{rd} \cdot Y_{rd} = E_{dd} \tag{9}$$

Subject to:

$$\begin{gathered} \sum_{r=1}^{s} \mu_{rd} \cdot Y_{rj} - \sum_{i=1}^{m} \omega_{id} \cdot X_{ij} \le 0, \ \forall j, \ j = 1,2,3,\ldots,n \\ \sum_{i=1}^{m} \omega_{id} \cdot X_{ij} = 1 \\ \omega_{id} \ge 0, \quad \forall i, \quad i = 1,2,3,\ldots,m \\ \mu_{rd} \ge 0, \quad \forall r, \quad r = 1,2,3,\ldots,s \end{gathered} \tag{10}$$

For each DMU$_\text{d}$ (d = 1, 2, 3, . . ., n), a group of optimal weights can be obtained by solving the CCR model in Equation (9). In the CCR model, each DMU is self-evaluated and termed efficient if the optimal objective function is equal to 1.

Table 1: Composition ratios of citronella oil compounds and their properties [1]

Citronella Oil Compounds	*Citronellal*	*Citronellyl Acetate*	*Geranial*	*Geranyl Acetate*	*Citronellol*	*Geraniol*
Formula	$C_{10}H_{18}O$	$C_{12}H_{22}O_2$	$C_{10}H_{16}O$	$C_{12}H_{20}O_2$	$C_{10}H_{20}O$	$C_{10}H_{18}O$
Composition ratio (based on mass)	0.259	0.016	0.159	0.032	0.076	0.458
Molecular weight, Mwi (g/mol)	154.25	198.3	152.23	196.29	156.27	154.25
Vapor pressure, P0 (pa)	2408.954	692.135	785.764	604.002	978.351	814.883

*The average density of citronella is 0.888 g/cm^3.

The decision variables, input and output variables, are energy cost and percentage oil yield, respectively. The objective function is to find the efficiency score (E_{dd}) of each DMU. The efficiency score is sought to be maximized, under the constraints that using those weights on each DMUd, no efficiency score exceeds one.

3 Materials and Methods

Many different essential oil extraction models are described in the literature. Among these models, the three-stage extraction model is one of the most efficient methods for obtaining the extracted oil, and it also predicts the extracted oil yield in various conditions related to steam flow rate and distillation time. The results from this model must be evaluated and ranked in terms of cost-effectiveness (between oil yield and energy cost) by using the DEA-CCR model. The framework for the proposed method is shown in Figure 2.

3.1 *Solving the three-stage extraction model for citronella oil*

Solving the problem of the three-stage extraction model includes the following steps:

3.1.1 Calculate the equilibrium concentration of citronella oils

The equilibrium concentration of citronella oils can be obtained from Equation (2) and the vapor pressure and molecular weight of citronella oil can be obtained from Equations (3) and (4), respectively.

3.1.2 Calculate the diffusivity and the mass transfer coefficient

The diffusivity and the mass transfer coefficient can be obtained from Equations (6) and (7), respectively.

3.1.3 Calculate the exudation kinetic constant and the oil spots average thickness

The exudation kinetic constant, K_{tr}, and the oil spots average thickness, h, were obtained using the nonlinear least square (curve-fitting) method [39], [40]. The least square algorithm was to choose the parameters that would minimize the deviations of the theoretical curve from the experimental points. This method was defined as follows [Equation (11)]:

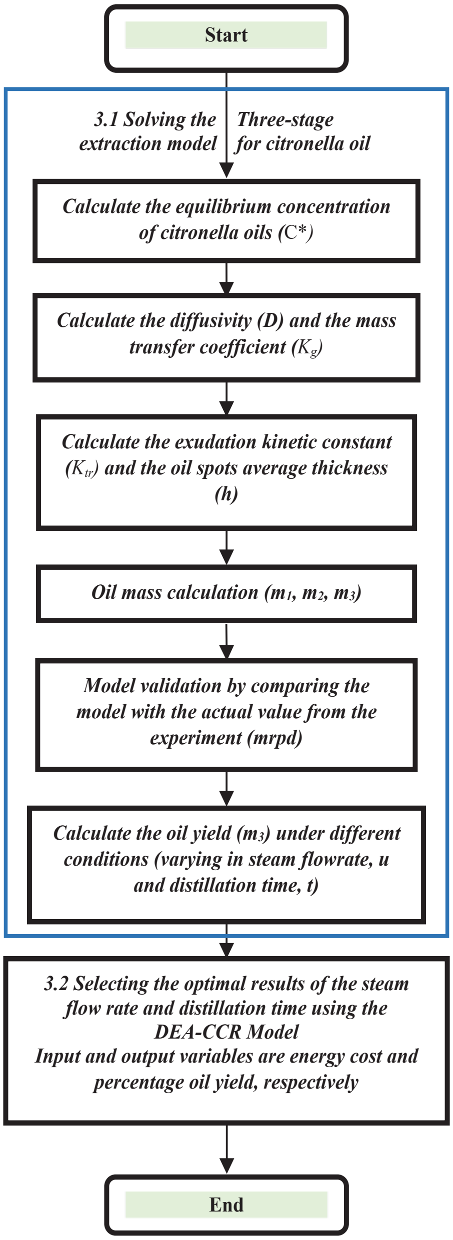

Figure 2: Framework for the proposed approach.

$$S = \min\left(\sum_{i=1}^{m}\left(y_i - F\left(x_i,\beta\right)\right)^2\right) \tag{11}$$

where x_i = the input data, y_i = the data obtained from the experiment, $\beta = (\beta_1, \beta_2, ..., \beta_n)$ = parameter vector to find the vector that best fits the curve of the given data with the least square sense. In this model, the parameters β_1 and β_2 are K_{tr} and h,, respectively. $F(x_i, \beta)$ is the system of differential functions can be obtained from Equations (1), (5), and (8).

The experimental data were obtained from the experiment in the laboratory. Steam was produced from the boiler and fed to the distillation pot. Inside the pot, the citronella leaves were cut into small pieces of about 1 cm, weighing 1,573 g, and placed on a sieve. The pressure was controlled at 20 psi and the distillation time was 180 min. Steam and essential oil were evaporated from the distillation pot and entered the condensing unit to separate the essential oils from the water.

3.1.4 Oil mass calculation

The values obtained from Section 3.1.1–3.1.3 were substituted in Equation (1), (5), and (8) to obtain a system of differential equations. Then it was further solved to obtain a value of oil mass in trichome, oil mass in the aqueous layer, and oil mass in condensing unit or oil recovery yield.

3.1.5 Model validation

In this step, the three-stage extraction model was compared with other models, pseudo-first-order model [26] and diffusional model [27]

In the pseudo-first-order model developed by Milojević *et al.* [26], the amount of oil yield was determined by the following Equation (12):

$$\frac{q}{q_\infty} = 1 - e^{-k_1 t} \tag{12}$$

where q = amount of essential oil, q_∞ = amount of essential oil distilled off until saturation, k_1 = the rate constants for diffusion processes in min^{-1}, t = time in min. This is the logarithmic equation based on the assumption of pseudo-first-order kinetics concerning the essential oil remaining in the plant material and is a frequently used model for both water and steam distillations. First-order kinetics have been used to model the essential oil extraction from various kinds of plants [41]–[43].

In the diffusional model developed by Cassel [27], [44], the steam distillation process was simulated using a model based on Fick's law in an unsteady state for one-dimensional rectangle geometry. The extracted mass of soluble constituent is presented as [Equation (13)]:

$$m(t) = \frac{8m_0}{\pi^2}\sum_{i=0}^{\infty}\frac{\left(1-e^{-(2i+1)^2\pi^2 k_2 t/l^2}\right)}{\left(2i+1\right)^2} \tag{13}$$

where l is the thickness of the plate in cm and k_2 is the effective diffusion coefficient in cm^2/min. The diffusional model was also used in the study by Boutekedjiret [45] and Benyoussef *et al.* [46].

The predicted oil recovery yield of each model was fitted with actual values obtained from the experiment. The result of the goodness of fit from each model was calculated using the mean relative percentage deviation (MRPD) between the predicted and actual values of the ratio, which is defined as follows [26]:

$$MRPD = \frac{100}{n}\sum_{i=1}^{n}\left|\frac{\left(\frac{q}{q_\infty}\right)_{pi}-\left(\frac{q}{q_\infty}\right)_{ai}}{\left(\frac{q}{q_\infty}\right)_{ai}}\right| \tag{14}$$

where subscripts p and a denote predicted and actual values, respectively, q = the percentage of oil yield.

3.1.6 Calculate the oil yield under different conditions

In this section, the results of oil recovery yield under different conditions were predicted. The main parameter of the calculation was the steam flow rate, which was varied between 5,000 and 60,000 cm^3/min and the distillation time was set at 180 min.

3.2 *Selecting the optimal results of the steam flow rate and distillation time using the DEA-CCR model*

The DEA-CCR model in Section 2.4 was used to calculate the efficiency scores of each DMU using the LINGO software. A higher value of efficiency score meant a higher ranking. The maximum value of efficiency scores was selected as the best solution, including the best value of steam flow rate and distillation time.

The data set of input (energy cost per oil yield) and output (percentage oil yield) variables were shown in Table 3. The efficiency scores were obtained using Equations (9) and (10). As a result, the ranking of all DMUs was obtained as listed in Table 3.

4 Results and Discussion

4.1 *The calculation results of the three-stage extraction model for citronella oil*

In this calculation, the operating conditions of the system were determined to be the same as the actual conditions to produce citronella oil in a case study of a factory in Yang Talat District, Kalasin Province. The system has a pressure value of 20 psig, the steam temperature is 127 °C, the steam flow rate was set at 50,000 cm^3/min. and sample weight of citronella leave was 1573 g. The calculation results are shown as follows.

4.1.1 Equilibrium concentration of citronella oils

The property values from Table 1 were taken into Equation (3) to calculate the vapor pressure of essential oils. The pressure of the citronella oil was found to be 4.282 kPa. The molecular weight M_w was obtained from Equation (4) as 2.634 g/mol. The vapor pressure of citronella oils and their molecular weights were taken into Equation (2). The concentration of citronella oils C* at equilibrium was 3.626 g/m^3

4.1.2 Diffusivity and the mass transfer coefficient

Diffusivity was calculated from Equation (7) resulting in 3.224 cm^2/min. From Equation (6), the mass transfer coefficient, K_g was 244.524 cm/min. with a bed porosity of 0.7, the steam volume flow rate was 50,000 cm^3/min, Re was 8.43 and Pe was 141.1.

4.1.3 Exudation kinetic constant and the oil spots average thickness

The values of Ktr and h were obtained using MATLAB software. The algorithm of nonlinear curve fitting [39], [40] and the solving of differential equation system [47]–[49] were used to solve this problem. The values of K_{tr} and h were obtained as 0.0318 min^{-1} and 18.68 μm.

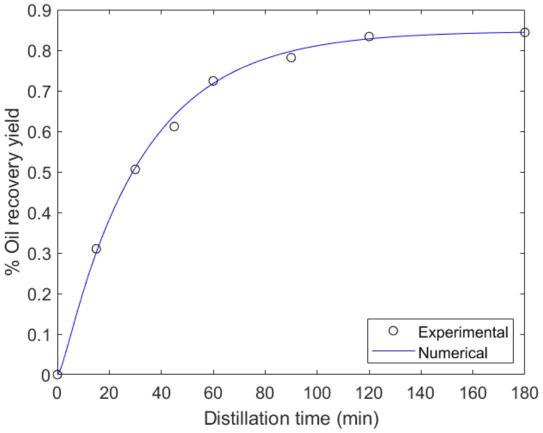

Figure 3: The percentage oil recovery yield.

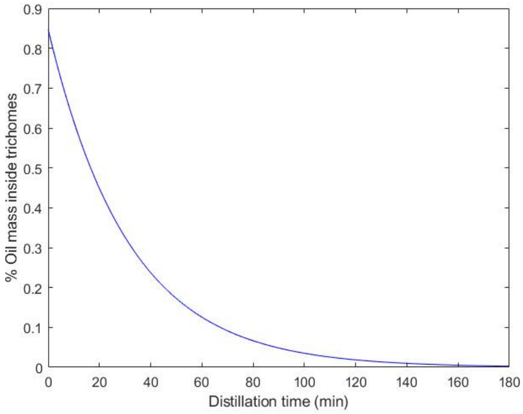

Figure 4: Graph between percent of oil mass inside trichomes and distillation time.

4.1.4 Oil mass calculation

The oil mass calculation results consisting of oil recovery yield, oil mass inside trichomes, and oil mass in the aqueous layer, were illustrated with graphs as in Figures 3–5, respectively.

In Figure 3, the percentage of oil recovery yield increased sharply during the first 80 min, after which the increase rate was slower and reached a maximum of 0.84% in 180 min.

To study the oil mass in the glandular trichome during the distillation period, the results were shown in Figure 4. The oil mass inside the trichomes decreased rapidly for the first 80 min and then gradually decreased until 180 min.

The oil mass in the aqueous layer during the

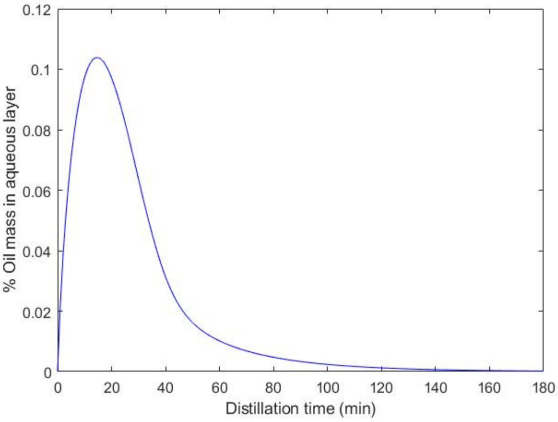

Figure 5: Graph between the percentage of the oil mass in the aqueous layer and distillation time.

distillation period was calculated from the model, and the results are shown in Figure 5. At the interface, the oil accumulated with a small amount, the maximum is 0.105% in the 17th min. After that, it decreased rapidly until the 80th min, and then slowly declined until the 180th min. In conclusion, there was little oil at the interface to accumulate, as most of the oil was rapidly forced by the steam to move into the condensation unit.

4.1.5 Model validation

In this section, the three-stage extraction model was compared with the results of the laboratory and other models. The index used for comparison was the mean relative percentage deviation (MRPD), as shown in Equation (14).

The results of plotting graphs between oil recovery yield at different distillation times compared between different models and laboratory experiments are shown in Figure 6. And the results of MRPD comparisons of the three-stage extraction model with pseudo-first-order model and diffusional model are shown in Table 2. The results expressed that the MRPD of the three-stage extraction model was only slightly greater than the pseudo-first-order model, but better than the value of the diffusional model.

Table 2: Comparions of the MRPD values from each model

	Three-Stage Extraction Model	Diffusional Model	Pseudo-first-order Model
MRPD	1.701	6.700	1.299

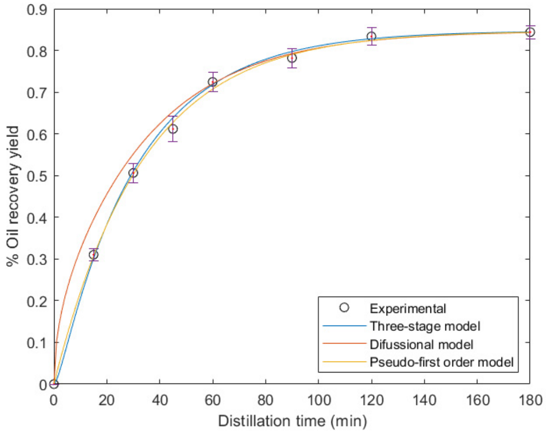

Figure 6: Models and experimental comparison of oil yield during the distillation period.

4.1.6 Calculate the oil yield under different conditions

The advantage of the three-stage extraction model was it was able to predict the yield of extracted oil which varies according to different conditions. The graph between stream volume flow rate and percentage of oil output is shown in Figure 7.

The graph shows that the different steam flow rates always affect the percentage of oil yield during distillation. It was analyzed by the quantitative classification of the steam flow rate as follows.

a) In the range of 55,000–60,000 cm^3/min, the k and l curves almost overlapped at every distillation period.

b) In the range of 40,000–50,000 cm^3/min, the h, i, and j curves reduced by 2–8% compared to a) in the first 80 min and then the curves change little.

c) In the range of 25,000–35,000 cm^3/min, the e, f, and g curves reduced by 16–40% compared to a) in the first 120 min and then the curves were not much changed.

d) When the steam flow rate was less than 20,000 cm^3/min, the a, b, c, and d curves decreased rapidly by over 50% compared to a) at every distillation period.

These results were applied to find optimal conditions for the distillation. If the distillation time or the steam flow rate was insufficient, a small amount of oil could be produced. Nonetheless, if the distillation time or the steam flow rate was too high, its energy would be wasted and its costs would be increased. Therefore, optimal conditions were selected to achieve optimal energy cost and oil yields, as shown in the next section.

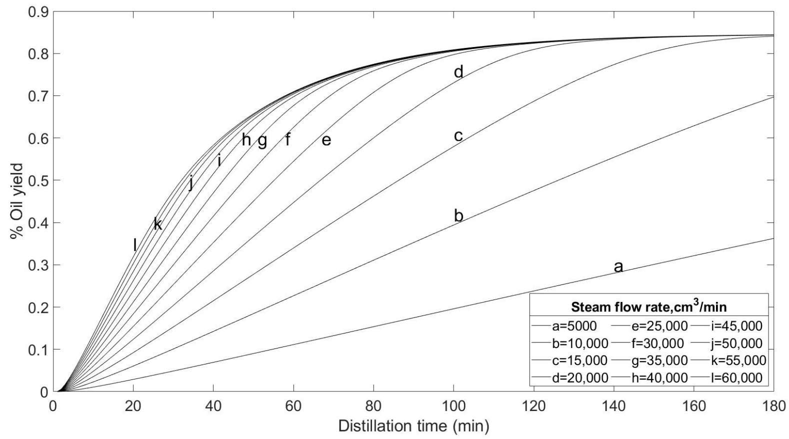

Figure 7: The graph between stream flow rate and percentage oil yield.

4.2 *Optimal results of the steam flow rate and distillation time using the DEA-CCR model*

The calculation results, including the percentage of oil yield, steam flow rate, and distillation time, were used to construct a decision matrix. In this matrix, the steam flow rate and distillation time were the DMUs, energy cost per oil yield (obtained from steam flow rate and time) was the input and the percentage of oil yield was the output. The decision matrix was evaluated using the DEA-CCR model. After that, all rankings of DMUs were obtained as shown in Table 3. For the first column of Table 3, the steam flow rate at different levels and at different distillation times was assigned in code for each DMU. For example, the code F5000T15 (DMU1) was meant a steam flow rate of 5,000 cm^3/min and at a time of extraction of 15 min.

Table 3: Ranking of citronella oil distillation conditions using the DEA-CCR model

Code	DMUs	Energy Cost Per Yield	Percent of Yield	Efficiency Score	Rank
F5000T15	DMU$_1$	7.803	0.021	0.002	84
F5000T30	DMU$_2$	3.291	0.051	0.010	82
F5000T45	DMU$_3$	2.771	0.082	0.019	80
F5000T60	DMU$_4$	2.192	0.110	0.032	78
F5000T90	DMU$_5$	1.843	0.180	0.061	75
F5000T120	DMU$_6$	1.715	0.240	0.088	71
F5000T180	DMU$_7$	1.302	0.360	0.174	61
F10000T15	DMU$_8$	3.844	0.045	0.007	83
F10000T30	DMU$_9$	1.579	0.110	0.044	77
F10000T45	DMU$_{10}$	1.385	0.170	0.077	73

Table 3: Ranking of citronella oil distillation conditions using the DEA-CCR model (*Continued*)

Code	DMUs	Energy Cost Per Yield	Percent of Yield	Efficiency Score	Rank
F10000T60	DMU$_{11}$	1.090	0.230	0.133	67
F10000T90	DMU$_{12}$	0.958	0.360	0.236	59
F10000T120	DMU$_{13}$	0.891	0.480	0.338	55
F10000T180	DMU$_{14}$	0.696	0.700	0.632	44
F15000T15	DMU$_{15}$	2.605	0.069	0.017	81
F15000T30	DMU$_{16}$	1.127	0.160	0.089	70
F15000T45	DMU$_{17}$	0.978	0.250	0.161	63
F15000T60	DMU$_{18}$	0.743	0.350	0.296	57
F15000T90	DMU$_{19}$	0.675	0.530	0.493	50
F15000T120	DMU$_{20}$	0.644	0.690	0.673	41
F15000T180	DMU$_{21}$	0.602	**0.840**	0.877	**16**
F20000T15	DMU$_{22}$	2.009	0.092	0.029	79
F20000T30	DMU$_{23}$	0.890	0.210	0.148	64
F20000T45	DMU$_{24}$	0.768	0.330	0.270	58
F20000T60	DMU$_{25}$	0.586	0.460	0.493	51
F20000T90	DMU$_{26}$	0.546	0.680	0.783	29
F20000T120	DMU$_{27}$	0.569	0.810	0.895	12
F20000T180	DMU$_{28}$	0.624	**0.840**	0.846	**20**
F25000T15	DMU$_{29}$	1.603	0.120	0.047	76
F25000T30	DMU$_{30}$	0.744	0.260	0.219	60
F25000T45	DMU$_{31}$	0.640	0.410	0.402	53
F25000T60	DMU$_{32}$	0.508	0.550	0.680	39
F25000T90	DMU$_{33}$	0.499	0.770	0.969	5
F25000T120	DMU$_{34}$	0.581	0.820	0.886	14
F25000T180	DMU$_{35}$	0.646	0.840	0.817	23
F30000T15	DMU$_{36}$	1.420	0.140	0.062	74
F30000T30	DMU$_{37}$	0.645	0.310	0.302	56
F30000T45	DMU$_{38}$	0.565	0.480	0.533	48
F30000T60	DMU$_{39}$	0.458	0.630	0.863	18

Table 3: Ranking of citronella oil distillation conditions using the DEA-CCR model (*Continued*)

Code	DMUs	Energy Cost Per Yield	Percent of Yield	Efficiency Score	Rank
F30000T90	DMU$_{40}$	0.503	0.790	0.987	3
F30000T120	DMU$_{41}$	0.594	0.830	0.878	15
F30000T180	DMU$_{42}$	0.668	0.840	0.790	25
F35000T15	DMU$_{43}$	1.284	0.160	0.078	72
F35000T30	DMU$_{44}$	0.574	0.360	0.394	54
F35000T45	DMU$_{45}$	0.519	0.540	0.654	43
F35000T60	DMU$_{46}$	0.439	0.680	0.974	4
F35000T90	DMU$_{47}$	0.519	0.790	0.955	7
F35000T120	DMU$_{48}$	0.613	0.830	0.850	19
F35000T180	DMU$_{49}$	0.690	0.840	0.765	32
F40000T15	DMU$_{50}$	1.178	0.180	0.096	69
F40000T30	DMU$_{51}$	0.533	0.400	0.471	52
F40000T45	DMU$_{52}$	0.498	0.580	0.731	36
F40000T60	**DMU$_{53}$**	**0.440**	**0.700**	**1.000**	**1**
F40000T90	DMU$_{54}$	0.536	0.790	0.926	9
F40000T120	DMU$_{55}$	0.633	0.830	0.824	22
F40000T180	DMU$_{56}$	**0.712**	**0.840**	0.741	**34**
F45000T15	DMU$_{57}$	1.093	0.200	0.115	68
F45000T30	DMU$_{58}$	0.511	0.430	0.528	49
F45000T45	DMU$_{59}$	0.489	0.610	0.784	28
F45000T60	DMU$_{60}$	0.447	0.710	0.998	2
F45000T90	DMU$_{61}$	0.553	0.790	0.898	11
F45000T120	DMU$_{62}$	0.653	0.830	0.799	24
F45000T180	DMU$_{63}$	**0.734**	**0.840**	0.719	**37**
F50000T15	DMU$_{64}$	1.023	0.220	0.135	66
F50000T30	DMU$_{65}$	0.492	0.460	0.587	47
F50000T45	DMU$_{66}$	0.495	0.620	0.786	27
F50000T60	DMU$_{67}$	0.460	0.710	0.969	6
F50000T90	DMU$_{68}$	0.562	0.800	0.894	13
F50000T120	DMU$_{69}$	0.672	0.830	0.776	30
F50000T180	DMU$_{70}$	0.756	0.840	0.698	38
F55000T15	DMU$_{71}$	1.007	0.230	0.143	65
F55000T30	DMU$_{72}$	0.496	0.470	0.595	46
F55000T45	DMU$_{73}$	0.502	0.630	0.789	26
F55000T60	DMU$_{74}$	0.473	0.710	0.941	8
F55000T90	DMU$_{75}$	0.579	0.800	0.869	17
F55000T120	DMU$_{76}$	0.692	0.830	0.754	33
F55000T180	DMU$_{77}$	0.778	0.840	0.678	40
F60000T15	DMU$_{78}$	0.953	0.250	0.165	62
F60000T30	DMU$_{79}$	0.489	0.490	0.629	45
F60000T45	DMU$_{80}$	0.516	0.630	0.767	31
F60000T60	DMU$_{81}$	0.487	0.710	0.916	10
F60000T90	DMU$_{82}$	0.595	0.800	0.845	21
F60000T120	DMU$_{83}$	0.711	0.830	0.733	35
F60000T180	DMU$_{84}$	0.800	0.840	0.659	42

From Table 3, the optimum conditions 1st rank from the study were at DMU53, code F400000T60, or at a steam flow rate of 40,000 cm^3/min and distillation time of 60 min, which an oil recovery yield at 0.7% and had an energy cost of 0.440 kWh/mL.

Although some ranks could provide an oil yield percentage higher than the optimum value above where the maximum oil yield from the table was 0.84%, the energy cost at those ranks provided much higher than the optimum value. For example, 34th and 37th ranks from Table 3 provided the maximum oil yield but used energy costs of 0.712 and 0.734 kWh/mL, respectively, it was presented almost twice as high as energy costs at optimum conditions (0.440 kWh/mL).

In addition, considering the use of steam flow rate, it was found that the 16th and 20th place both also provided maximum oil yields, but the use of steam flow rates was as low as 15,000 and 20,000 cm^3/min, respectively. Using a low steam flow rate would lead to a longer distillation time (Figure 7) and might affect the quality of the oil extracted. Therefore, the optimal value was the 1st rank from this study which provided the low energy cost, and a steam flow rate of 40,000 cm^3/min which was a flow rate that maintained good oil quality.

When comparing the optimization results with the actual values obtained from laboratory experiments, it was found that the average oil recovery yield obtained from the experiment was 0.706% and the cost of energy used was 0.444 kWh/mL. Therefore, the error of computation compared to the actual test results was equal to 0.85% for oil yield and 0.98% for energy cost, respectively, which was within acceptable limits. Moreover, when the actual experiment was performed under the calculated optimum conditions and compared with the traditional operating conditions it turned out that the energy consumption was reduced by 34.6%.

In terms of cost-effectiveness, the energy cost of citronella oil extraction by steam distillation was similar to a supercritical fluid and solvent extraction method [10]. Although the percentage of oil yield was about 10% that less than both extraction methods [10], the steam distillation method was probably more cost-effective because both extraction methods provided the high machine cost of the supercritical fluid method and an additional cost of oil purification of the solvent extraction method.

In addition, many studies apply heat-assisted technology to the extraction of essential oils, such as ohmic-heated hydro-distillation [50] and microwave-assisted steam distillation [51]. Both methods can reduce the cost of energy compared to steam distillation with 40–60% lower costs. Considering the limitations of citronella oil extraction by steam distillation from this study, the optimum oil yield of 0.7% was relatively low. An alternative extraction method with low energy costs but high oil yield is the microwave technique [51]. In future studies, microwave energy to assist in the heating method will be applied. It is expected to provide consistent heating as well as to save energy and increase productivity.

The three-stage extraction model was validated by comparing the output to experimental data sets that aligned with the simulated scenario. The MRPD value of this model was also compared with the pseudo-first-order model [26] and the diffusional model [27], which were derived from the kinetic equation and Fick's law equation, respectively. These models [41]–[43], [45], [46], [52] have been widely used in oil extraction analysis. According to the compared results, it was found that the calculation with the three-stage extraction model compared to the experimental results presented a good MRPD error of 1.7%, which was similar to the pseudo-first-order model, but much better than the diffusional model, which had a value of 6.7%. In addition, when comparing the predicted values of oil yield in an optimal condition of the three-stage extraction model with the experimental results, there was a slight deviation of 0.85%.

Considering the advantages of a three-stage extraction model, it was found that the model could predict oil yields at different steam flow rates and distillation times. Prediction results had several options which were evaluated using DEA-CCR techniques to determine the optimal values. This method differed from previous studies that used optimization methods based on experimental results which were time-consuming and costly [21]–[23]. In addition, the three-stage extraction and DEA-CCR model was also used to calculate the optimal yield and energy cost for other essential oils [28],[29]. In contrast, the DEA-CCR technique presented the limitation that it had only one of the DMUs, which was the maximum efficiency score (Edd =1). However, in other cases, if more than one DMU has the maximum efficiency score, other techniques would be considered. The techniques such as DEA cross-efficiency and common-weights might be an option that can solve this problem [53], [54].

5 Conclusions

This research studied the citronella oil extraction by steam distillation with the calculation method using a three-stage extraction model. This study was beneficial as it could be applied to calculate the change in oil mass inside trichomes, oil mass in the aqueous layer, and oil recovery yield. In addition, this study was also apply to predict oil yield at various steam flow rates and distillation times. The model validation illustrated that the three-stage extraction model had reliable predictive accuracy. The DEA-CCR model was also combined in this research to assess factors including energy cost and oil yield and to determine the optimum value. This hybrid method was a new method that presented the advantage of obtaining optimum conditions by calculation technique. It could reduce the time and resources used in laboratory experiments. The results were performed at the optimum conditions compared to the traditional operating conditions; it achieved a 34.6% reduction in energy consumption. When considering together with the cost of energy, it presented a relatively low optimum oil yield of 0.7%. Therefore, in future studies, the researcher might combine microwave technology with the traditional heating method (gas firing) to help increase productivity while maintaining low energy costs.

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