STUDY ON MATHEMATICAL MODEL IN SIMULATING CYMBOPOGON WINTERIANUS ESSENTIAL OIL EXTRACTION BY STEAM DISTILLATION

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Abstract

The main objective of this study is to improve the mathematical modelling of Cymbopogon winterianus essential oil extraction by steam distillation proposed by Cassel and Vargas by minimum 5% error reduction. Two process variable of steam distillation which are extraction time and raw material state (dry or natural) has been optimized by using factorial experimental planning to obtain high yields of citronella essential oil from twig and leaves of lemongrass species Cymbopogon winterianus (C.winterianus). The optimal condition for maximum yield (0.942%) were found to be an extraction time, 4 hr, state, natural plant. The study of Cassel and Vargas was subsequently continued with five proposed kinetics model of the extraction process. The modelling of the extraction process is optimized by using one adjustable parameter of the model and the adequacy of the fit of the models to the experimental data are analyzed by using three statistical criteria that are correlation coefficient (r) ,the root mean square error (RMSE) and the mean relative deviation modulus (E). The result has shown that the mathematical model developed by Ana based on mass transfer fundamentals is the optimum mathematical model for the extraction of Cymbopogon winterianus essential oil by steam distillation.

Keywords: Essential Oils, Cymbopogon winterianus essential oil, Optimization of mathematical model of extraction of citronella essential oils

1. Introduction

Cymbopogon winterianus is commonly known as Citronella. Citronella Grass and Java Citronella grass is a lemongrass species that is believed to have originated from Cymbopogon nardus that often referred to Ceylonese, a Sri Lankan commercial citronella. Cymbopogon winterianus was named after the Winter whom presented the plant as a separate species in 19th century. It is later than introduced to Indonesia and commercially known as Javanese citronella. The plant later was further introduced to India in 1959 [1].

Essential oils are subtle, natural, aromatic and volatile compounds which are extracted from the flower, seeds, leaves, stems, bark and roots of herbs [2]. As agreed by Tajidin et al (2012), essential oils are natural products which can be extracted from plants. They were formed through mixture of varied and complex volatile chemical compounds with high proportion of terpene

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associated with aldehyde, ketone and alcohols. The different mixtures of the chemical then were deposited in different structure of the plant [3]

Essential oil can be extracted traditionally by hydrodistillation, steam distillation, or solvent extraction [4]. Kabuba (2013) also stated that essential oils derived from aromatic plant are typically extracted by steam distillation as this method is simple and relatively inexpensive process. The essential oil in the plant is removed by steam of water vapour and can be separate easily in the next phase [5]. The aromatic industry often employed this extraction method as it is cheaper than advanced methods such as supercritical fluid extraction (SFE). The proportion of different essential oil extracted by steam distillation in the industry is 93% and the remaining 7% employed other extraction methods such as SFE and microwaves extraction [5]. This method is also preferred by Amenaghawon et al (2014) because steam distillation is flexible, versatile, do not lead to decomposition of essential oils and provides the possibility of operating with small volumes [6].

Essential oils played an important role in the personal and social hygiene of mankind in terms of their application in cosmetics, toiletries, medicinal formulation, surface coatings and aroma therapy [7]. Essential oils have been largely applied in various areas due to their natural properties which exhibit antibacterial, antifungal, insecticidal, antiviral and anti-herbivore characteristics. To date, approximately 3000 essential oils are known at the and 200 of the essential oils are commercially important especially in pharmaceutical, agronomic, sanitary, food, cosmetics and perfume industries [4].

Citronella oil is a clear liquid that possesses strong lemon-like odour and pale yellow colour. The total amount of essential oils present in lemongrass varies between 0.28% and 1.4%. Maximum recovered recorded is at 3% [6]. Citronella oil can be fractionated into three major constituent chemicals which are citronellal, citronelol and geraniol. The three major constituent chemicals are used commercially in the industry for many purposes such as soaps, sprays, disinfectants and polishes [1]. The major constituent of the essential oil is citral which is more than 75% by weight of the essential oil [4]. Citral is the natural combination of two isomeric aldehydes namely isomers geranial (α-citral) and neral (β-citral). Other unusual active components are limonene, geraniol, citronella and β-myrcene [3]. The essential oil quality is judged by its citral content and the oil solubility in alcohol. The citronella essential oil can be in reddish-yellow to reddish brown colour with strong lemon odour properties. It is used in perfume, cosmetics and soap. Citronella essential oil serves as important raw material to pharmaceutical preparation such as pain balm, disinfectants and mosquito-repellent creams [4].

Mathematical modelling of steam distillation is considered an inevitable step to project industrial plants that have good operational conditions. The advantage of using mathematical models is the process can be simulated without running the experimental procedures in order to know the extraction process behaviour. Therefore the mathematical model allows alternative strategies to be tested in order to evaluate the selection of the process variable conditions [9]. Another advantage of having a well-accepted mathematical model is its usefulness in the development of up-scaling procedures from laboratory to pilot and then to industrial scale [5]. By modelling the kinetics of distillation or any other process, it can contribute not only toward the fundamental understanding of the process but can also contribute towards better control and higher efficiency [10].

Mathematical modelling is flexible as it can be varied from simple mathematical model to complex mathematical model. Many researchers construct their own mathematical model by using various physical laws with the aid of mathematical software such as Matlab, ANOVA and Designer Software 6.0.6. Benyoussef et al (2002) modelled the steam distillation of essential oil from coriander by using two diffusion models that take into account both diffusion and transfer of species [11] whereas Cassel and Vargas modelled the steam distillation of lemon grass using a model based on Fick's law steady state for one-dimensional rectangular geometry [12]. On the other hand, Ha et al modelled the extraction of essential oil from lemon grass stems by using two-factor interaction model and a linear mode in terms of coded variables for the extraction of essential oil from lemon grass stem [13].

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2. Laboratory experiment

In the previous research done by Cassel and Vargas (2006) in their research "*Experiments and Modelling of the Cymbopogon winterianus Essential Oil Extraction by Steam Distillation*", a laboratory experiments were conducted to obtain a real data on the extraction of the Cymbopogon winterianus essential oil. The data obtained from this laboratory scale experiment were used as a benchmark and reference in developing the mathematical model of the process [12].

2.1 Material and method

The experiment was conducted by using 0.04kg of citronella leaves samples with the average thickness of 5.25x10-4m. The total extraction time is 114 min with the maximum yield of 1.02 g of essential oil. The principal compounds of citronella essential oil found are citronellal, citronellole, geraniole, geranyl acetate and α-cadinol. The experimental data from steam distillation for citronella is shown in Table 1 [12].

2.2 Degree of extraction

The degree of extraction, e is found by using the experimental data obtained and the mathematical formula (1) [12].

$$e(t) = \frac{m_A(t)}{m_{A(\infty)}} \tag{1}$$

Where $e(t)$ is the degree of extraction time at time t, $m_A(t)$ is the mass of citronella essential oil in gram unit extracted at time t and $m_A(\infty)$ is the maximum mass of citronella essential oil in gram unit extracted at the end of the experiment. The value of $e(t)$ calculated is shown in Table 1.

2.3 Yield of citronella essential oil

The yield of the extracted citronella essential oil is calculated by using the experimental data obtained and the mathematical formula (2) [12].

$$\text{Yield} = \frac{m_A(t)}{m_s} \tag{2}$$

Where $m_A(t)$ is the mass of citronella essential oil in gram unit extracted at time t and m_S is the mass of the citronella leaves sample in gram unit used at the beginning of the experiment. The extracted citronella essential oil yield calculated is shown in Table 1.

3. Kinetic model proposed

Models derived from Fick's second law of diffusion are frequently used to describe the kinetics of the extraction of different constituents from plant material. Simplified model and second-order kinetic model is also often used depending on the type of extraction method used and plant material itself [14]. A physical model that is derived based on simultaneous washing and diffusion to describe the system kinetics of essential oil extraction is often known second order kinetics while simplified model that assume either instantaneous washing followed by diffusion or diffusion with no washing is known as first order kinetics [15]. The main goal is to compare the first-order kinetic model

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and second-order kinetics model and suggest the most optimum mathematical model for Cymbopogon winterianus Essential Oil Extraction by Steam Distillation.

3.1 Cassel and Vargas (2006) method

In the research by Cassel and Vargas (2006) "*Experiments and Modeling of the Cymbopogon winterianus Essential Oil Extraction by Steam Distillation*", a mathematical model based on Fick's law in steady-state for one-dimensional geometry was developed and used to predict the yield of citronella essential oil extracted.

In the mathematical model, the assumptions made are:

1. At the beginning of the process the soluble constituent's concentration is homogenous and constant for the entire particle in the plant.
2. The steam carries out all the essential oil at the boundaries thus making the oil concentration at the boundaries very small ($c_A=0$).

The mathematical formulation is express as in (3) shown below [12].

$$\frac{\partial^2 c_A}{\partial x^2} = \frac{1}{D}\frac{\partial c_A}{\partial t} \text{ in } 0 \leq x \leq L \tag{3}$$

With the initial condition stated as in (4) [12]

$$c_A = c_{AO} \text{ in } t = 0 \tag{4}$$

And the boundary conditions showed as in (5) and (6) [12]

$$c_a = 0 \text{ in } x = 0 \tag{5}$$
$$c_a = 0 \text{ in } x = L \tag{6}$$

Where c_A is the concentration at time t, c_{AO} is the initial concentration at time $t=0$, L is the thickness of the plant leaves ad D is the effective diffusion coefficient. By using separation variable technique the following solution (7) till (8) was established [12].

$$c_A(x,t) = \sum_{n=1}^{\infty} \frac{4c_{A0}}{n\pi} \text{ sen } (\beta_n x) e^{-D\beta_n^2 t} \tag{7}$$

$$\beta_n = \frac{n\pi}{L} \tag{8}$$

With $n=1, 2, 3$….The mass flow as function of time was obtained from mass flux at boundary multiplied by the normal surface area which resulting in (9) [12].

$$\dot{m}_A(t) = \frac{4C_{A0}DA}{L} \sum_{m=0}^{\infty} e^{-D\beta_n^2 t} \tag{9}$$

Therefore, the extracted mass of soluble constituent is shown in (10) [12].

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$$m_A(t) = \frac{8m_{A0}}{\mu^2}\sum_{m=0}^{\infty}\frac{(1-e^{-\frac{(2m+1)^2\pi^2 Dt}{L^2}})}{(2m+1)^2} \tag{10}$$

The degree of extraction is obtained as shown in (11) [12].

$$e(t) = \frac{m_A(t)}{m_\infty} = \frac{\sum_{m=0}^{\infty}\frac{(1-e^{-\frac{(2m+1)^2\pi^2 Dt}{L^2}})}{(2m+1)^2}}{\sum_{m=0}^{\infty}\frac{1}{(2m+1)^2}} \tag{11}$$

3.2 Hervas et al (2006) method

Hervas *et al* (2006) proposed a first-order kinetic mechanism shown as in (12) to study the extraction process under the equilibrium condition [2].The model is also known as pseudo-first order kinetics where no washing of the essential oil occurs. The pseudo-first order kinetics is a logarithmic equation that is frequently used to model both water and steam distillations [15].

$$\frac{dC}{dt} = k(C_o - C) \tag{12}$$

Where C is the amount of essential oil produced, *t* is the extraction time in hours, C_o is the initial essential oil present and k is the effective diffusion coefficient. (12) is then integrated between the initial moment and a given point at time *t* to (13) [16].

$$C = C_0(1 - e^{-kt}) \tag{13}$$

The simulation of the model is run by using Microsoft Excel programming and the optimum value *k* is found by using the minimization of the root mean square error (*RMSE*) shown as in (18).

3.3 Garkal et al (2012) method

Garkal *et al* (2012) proposed a second order rate with the k is the second-order extraction rate constant. The mathematical model is shown as in (14) [17]. The mathematical model is integrated with the initial boundary condition, *t=0* to *t* and *Ct=0* to *Ct* shown as in (15) [17].

$$\frac{dC_t}{dt} = k(C_s - C_t)^2 \tag{14}$$

$$\frac{C_t}{t} = \frac{1}{\left[\frac{1}{k}C_s^2\right] + \left[\frac{t}{C_s}\right]} \tag{15}$$

Where *Ct* is the mass of oil extracted at time *t*, *t* is the extraction time in *Cs* is the initial amount of mass in the plant that is assumed to be the maximum mass of essential oil extracted over time. The simulation of the model is run by using Microsoft Excel programming and the optimum value *k* is found by using the minimization of the root mean square error (*RMSE*) shown as in (18).

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3.4 Ana et al (2007) method

Ana *et al* (2007) proposed a second order rate with two k parameter which each represents the washing and diffusion separately. The mathematical model is shown as in (16) where *C(t)* is the mass of oil produce at time *t*, *t* is the extraction time in hours, *Co* is the initial mass oil of the essential oil at time *t=0* which usually assumed to be zero, *K1* is the extraction rate constant and *K2* is the extraction capacity constant [18].

$$C(t) = C_o + \frac{t}{K_1 + K_2 t} \quad (16)$$

The simulation of the model is run by using Microsoft Excel programming and the optimum value *K1* and *K2* are found by using the minimization of the root mean square error (*RMSE*) shown as in (18).

3.5 Milojevi et al (2008) method

Milojevi *et al* (2008) proposed a second order rate with the k is the second-order extraction rate constant. The mathematical model is shown as in (17). The mathematical model is derived using Fick's second law at unsteady state for one-dimensional rectangle geometry [19].

$$\frac{q_o - q}{q_o} = A.\,e^{-kt} \quad (17)$$

Where *qo* represents the essential oil present in the citronella leaves which is assumed to be the maximum oil extracted in the experimental data, *q* is the essential oil produce at time *t*, *A* is a constant and *K* is the kinetic constant that includes the diffusion coefficient. The simulation of the model is run by using Microsoft Excel programming and the optimum value *A* and *k* are found by using the minimization of the root mean square error (*RMSE*) shown as in (18).

3.6 Statistical criteria used

The adequacy of the fit of the models to the experimental data are analyzed by using three statistical criteria that are correlation coefficient (*r*) shown as in (18) [20], the root mean square error (*RMSE*) shown in (19) [14], and the mean relative deviation modulus (*E*) shown in (20) [14] as the application of only one statistical criteria use for observation is not sufficient in evaluating non-linear mathematical model [21]. The model fits the experimental data well when *RMSE*→0, *r*→1 and *E*‹10% [14].

$$RMSE = \sqrt{\frac{1}{N}\sum_{i=1}^{N}\left(M_{experiment,i} - M_{predicted,i}\right)^2} \quad (18)$$

$$r = \frac{n\sum xy - \sum x \sum y}{\sqrt{\left[n(\sum x^2) - (\sum x)^2\right]\left[n(\sum y^2) - (\sum y)^2\right]}} \quad (19)$$

$$E = \frac{100}{N}\sum_{i=1}^{N}\left|\frac{C_{experiment,i} - C_{predicted,i}}{C_{experiment,i}}\right| [\%] \quad (20)$$

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4. Result and discussion

4.1 Laboratory result

The experimental data from steam distillation for citronella is shown in Table 1 and the yield of the essential oil extraction as time function is shown in Figure 1 [12].

Table 1. Experimental data from steam distillation for citronella as time function [12].

Time Extraction (min)	Mass Oil Extracted (g)	Degree of Extraction, e	Yield
0.0	0.0000	0.0000	0.0000
3.0	0.0850	0.0833	0.0021
6.0	0.1700	0.1667	0.0043
9.0	0.3400	0.3333	0.0085
12.0	0.4250	0.4167	0.0106
16.5	0.5100	0.5000	0.0128
21.0	0.5950	0.5833	0.0149
25.5	0.6800	0.6667	0.0170
36.0	0.7650	0.7500	0.0191
66.0	0.8500	0.8333	0.0213
90.0	0.9350	0.9167	0.0234
114.0	1.0200	1.0000	0.0255

It can be seen that in between zero minute to 25 minute the extraction of the citronella essential oil happen rapidly as the gradient of the graph at the early stage is almost linear increase in the cumulative volume of essential oil with time. The extraction of the citronella essential oil started to slows down after 25 minute and increases gradually until 120minute. Although the extraction of the citronella essential oil increases rapidly during the first 25 minutes, the extraction is not in linear and uniformly order. This is probably due to the variations in the heat supplied to the water boiler thus affecting the amount of steam supply to the extraction chamber.

By the increases of extraction time, the amount of oil extracted also increases until 0.019125 g/g yield at 36 minutes. Further increase in the extraction time result in not significant increases in the amount of oil extracted. Through the experimental result obtained, it is confirm that the kinetics of the diffusion in extraction of citronella essential oil obeys the Fick's second law of diffusion which follows the pattern after a certain time, the solute concentration in the citronella and the steam solvent achieve final equilibrium [22].

Thus the extraction of the citronella essential oil can be divided into two processes that are washing process and dissolve process. Washing phase refers to the fast phase where initial increase is caused by quick dissolution of easily accessible solute from the plant cell into the steam solvent [14]. The increase in temperature due to the steam supplied to the plant increases the pressure within the plant cell containing the essential oil. When the pressure reached certain level, the cell membrane breaks thus releasing the essential oil [15].

The washing process occurs when the external cell on the plant surface start to break thus releasing the essential oil. This process occurs almost instantaneously and in fast period of time. The essential oil from the internal cell of the plant must diffuse from the interior of the plant particle to the plant external surface. This process known as diffusion process where it occurs more slowly after the washing phase has completed [15].

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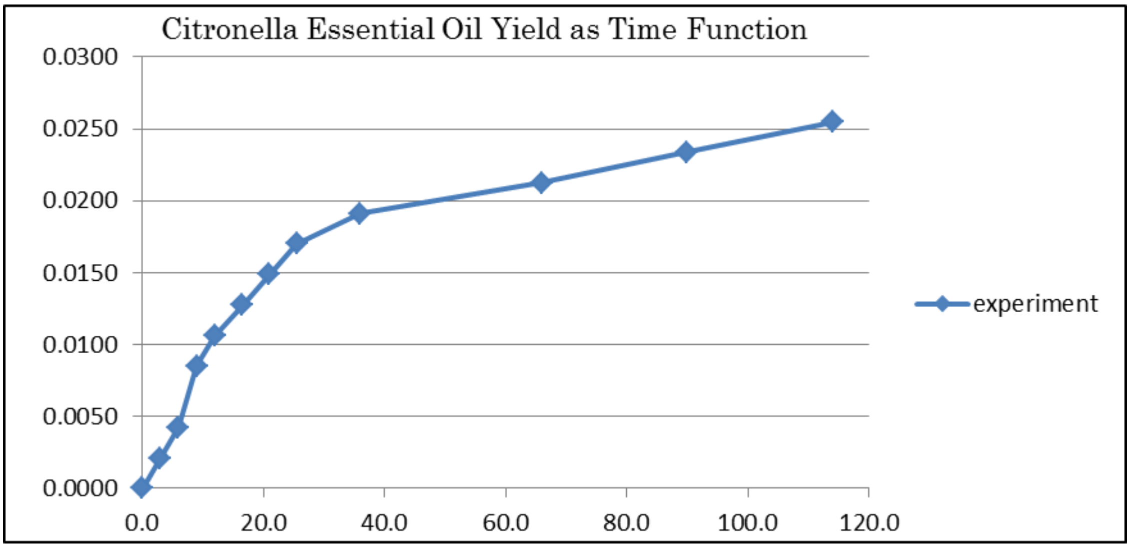

Figure 1: Citronella experimental yield as time function (Cassel and Vargas, 2006).

4.2 Cassel and Vargas (2006) method

Cassel and Vargas (2006) proposed optimum parameter *D* 1.60×10^{-11}. This parameter was estimated using the minimization of sum of square of errors between the experimental data and the prediction from the model. The minimum value is found by using Nelder- Mead Simplex method with software Matlab version 6. The result of the simulation is shown in Figure 2.

From Figure 2 shown, it can be clearly seen that the mathematical model and parameter *D* value proposed by Cassel and Vargas (2006) does not have the perfect fit in predicting the citronella essential oil yield extracted over time. The *RMSE* error calculated is 31.79% showing the differences between the experimental result and the prediction made by the mathematical model. This probably due to the assumption made at the early stage where parameter *D* is assumed to be constant overtime whereby in the experiment conducted in the laboratory scale, two process in the extraction was identified that are washing process and diffusion process. The mathematical model also does not take into account the resistance occurs in diffusion of essential oil from the interior of the plant to the external surface of the plant.

The mathematical model proposed by Cassel and Vargas (2006) is further optimized by finding the optimum parameter *D* value. The new optimum value of parameter *D* is found by using the minimization of the root mean square error (RMSE) shown in (18). The new optimum value of parameter *D* found by using the solver is 5.4×10^{7}. The predicted mass essential oil extracted and yield is shown in Figure 3.

By comparing the data on Figure 2 and Figure 3, we can observe a significant improvement in the mathematical model with the reduction of *RMSE* error from 31.79% to 22.61% after the parameter *D* is optimized. Nevertheless, the mathematical model still failed to predict the extraction of the essential oil after the washing stage and does not fit the experimental data perfectly. This confirms the assumption on the error made earlier where parameter *D* is assumed to be constant overtime does not take into account the resistance occurs in diffusion of essential oil from the interior of the plant to the external surface of the plant thus limiting the ability of the mathematical model to predict the amount of citronella oil extracted over time.

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4.3 Hervas *et al* (2006) method

The simulation of the model is shown in Figure 4 and the optimum value *k* (0.04) is found by using the minimization of the root mean square error (*RMSE*) shown in (18). Correlation coefficient (*r*) (0.3602) and the mean relative deviation modulus (*E*) (4.5204) are found using (19) and (20).

4.4 Garkal *et al* (2012) method

The simulation of the model is shown in Figure 5 and the optimum value *k* (0.06292) is found by using the minimization of the root mean square error (*RMSE*) shown in (18) .Correlation coefficient (*r*) (09923) and the mean relative deviation modulus (*E*) (17.92) are found using (19) and (20).

4.5 Ana *et al* (2007) method

The simulation of the model is shown in Figure 6 and the optimum value *K1* (19.09) and *K2* (0.8301) are found by using the minimization of the root mean square error (*RMSE*) shown in (18). Correlation coefficient (*r*) (0.9943) and the mean relative deviation modulus (*E*)(11.5183) are found using (19) and (20).

4.6 Milojevic *et al* (2007) method

The simulation of the model is run shown in Figure 7 and the optimum value *A* (1.003) and *k* (0.0402) are found by using the minimization of the root mean square error (*RMSE*) shown in (18). Correlation coefficient (*r*) (0.9931) and the mean relative deviation modulus (*E*) (9.0641) are found using (19) and (2

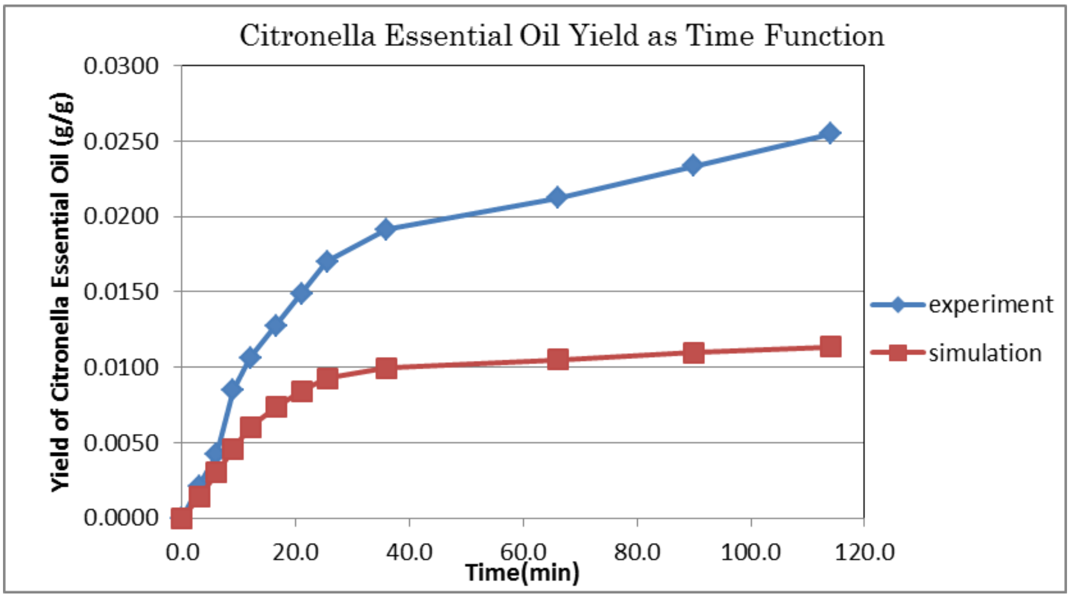

Figure 2. Cassel and Vargas citronella essential oil predicted yield as time function

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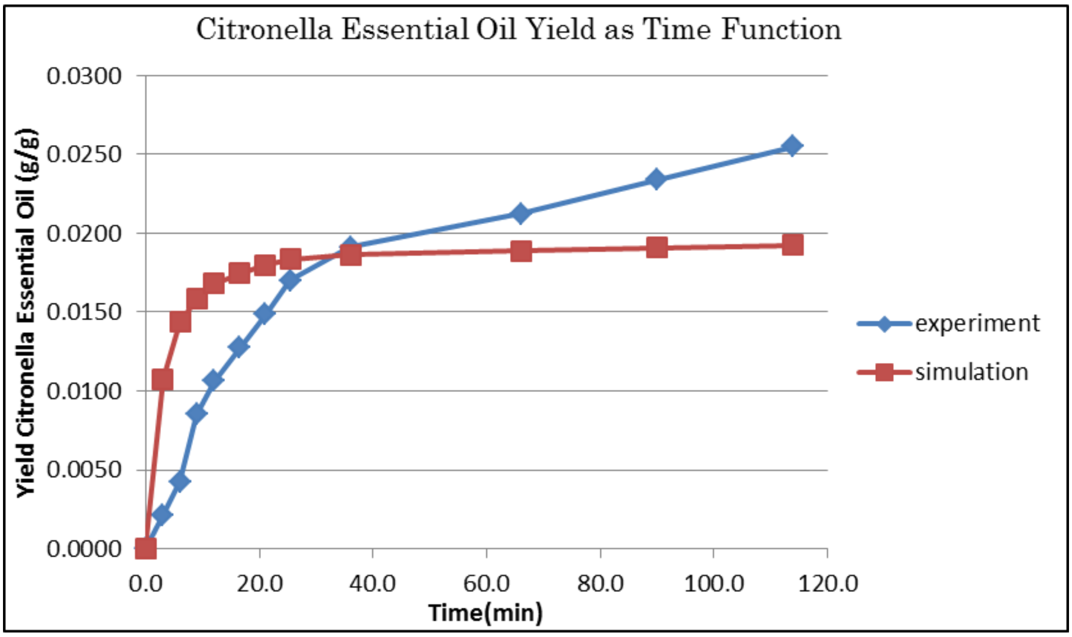

Figure 3. Optimized Cassel and Vargas citronella essential oil predicted yield as time function

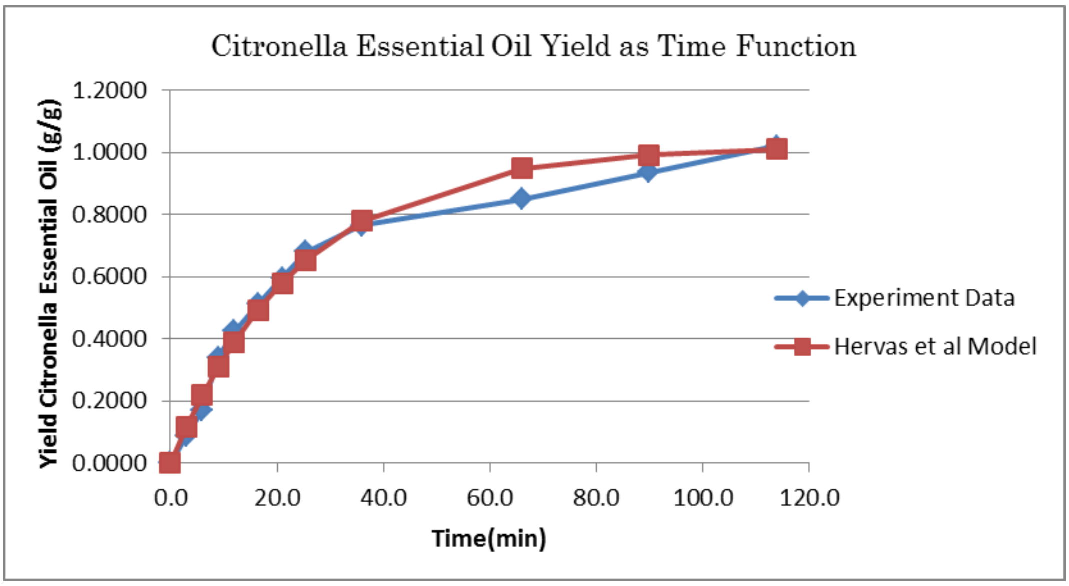

Figure 4. Hervas *et al* (2006) mathematical model predicted mass oil extracted as time function

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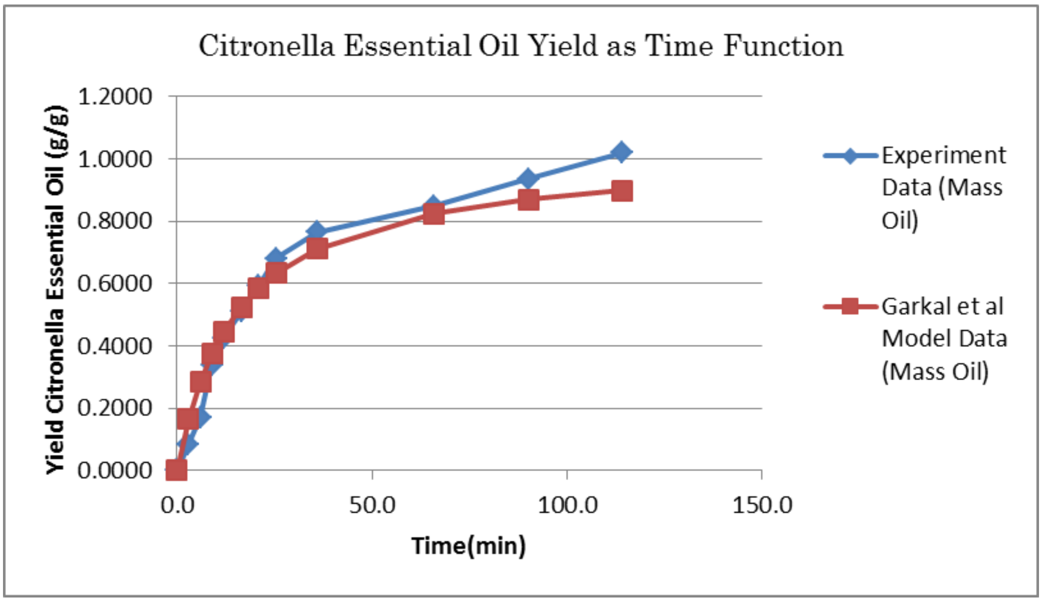

Figure 5. Garkal *et al* (2012) mathematical model predicted mass oil extracted a as time function

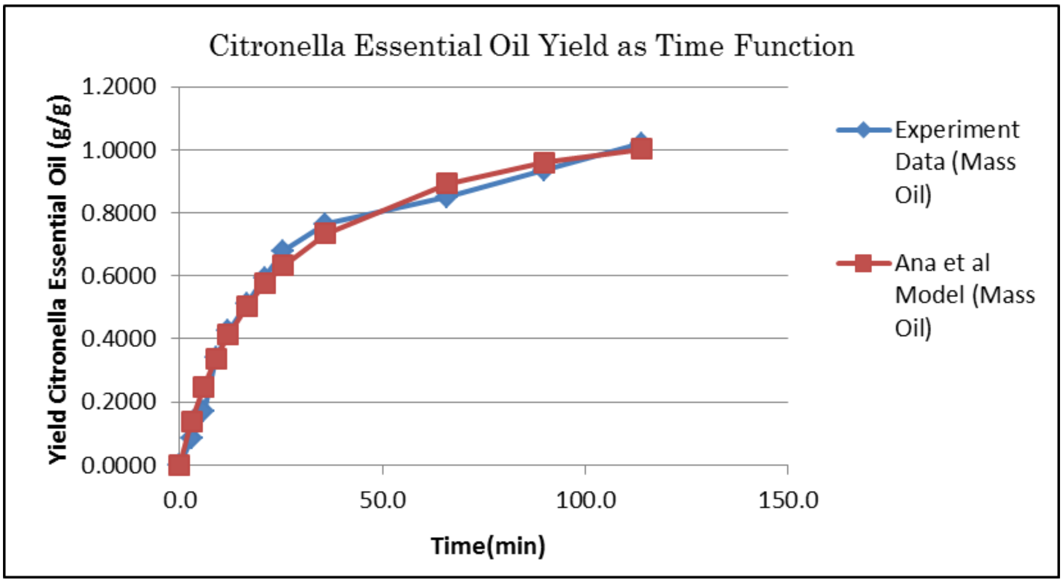

Figure 6. Ana *et al* (2007) mathematical model predicted mass oil extracted as time function

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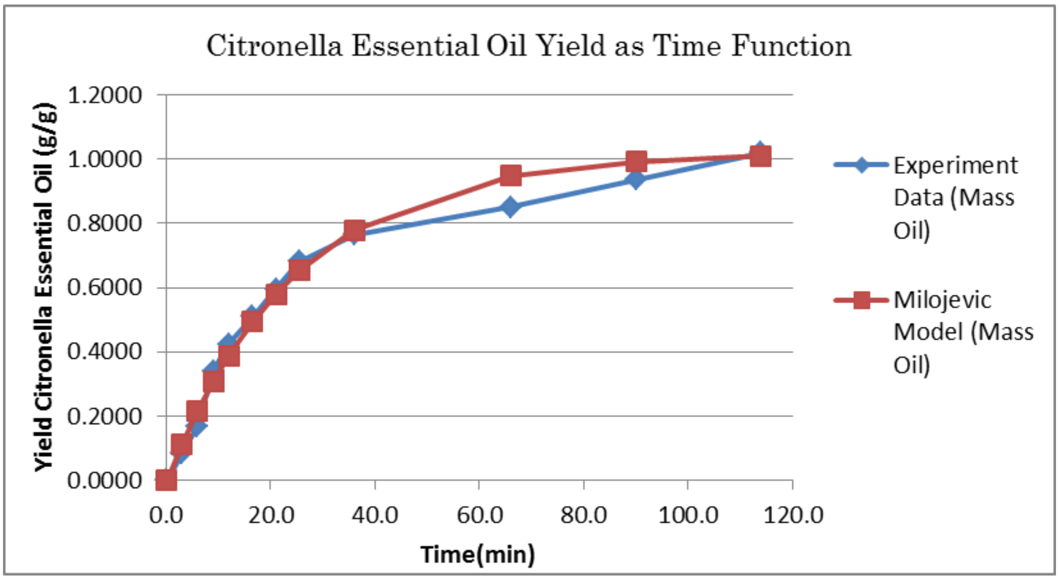

Figure 7. Milojevic *et al* (2008) Mathematical Model Predicted Mass Oil Extracted as Time Function

4.7 Statistical criteria result

The proposed mathematical model and parameter *D* value by Cassel and Vargas (2006) does not have the perfect fit in predicting the citronella essential oil yield extracted over time. The *RMSE* error calculated for the mathematical model and parameter *D* value proposed is 0.317916 (31.79% error). By optimizing the value of parameter *D* by using the minimization of the root mean square error (*RMSE*) as shown in (18) and Microsoft Excel Solver, the new optimum parameter *D* value is $5.4x\ 10^7$. Nevertheless the *RSME* error calculated is still very high 0.226 (22.6% error).

Table 2. Statistical Criteria Result

Model	RMSE	RMSE (%)	r	E (%)
Hervas *et al* (2006)	0.0424	4.2435	0.3602	4.5204
Garkal *et al*(2012)	0.0647	6.4662	0.9923	17.9200
Ana *et al* (2007)	0.0377	3.7713	0.9943	11.5183
Milojevi *et al* (2008)	0.0424	4.2414	0.9931	9.0641
Best Model	Ana *et al* (2007)	Ana *et al* (2007)	Ana *et al* (2007)	Hervas *et al* (2006

Thus to find the most optimum mathematical model for Cymbopogon *winterianus* Essential Oil Extraction by Steam Distillation, four mathematical models consist of first order kinetic and second order kinetic is studied. The mathematical models being studied are mathematical model proposed by Hervas *et al* (2006) Method, Garkal *et al* (2012) Method, Ana *et al* (2007) Method and Milojevi *et al* (2008) Method. Three criteria that are correlation coefficient (*r*), the root mean square error (*RMSE*) and the mean relative deviation modulus (*E*) is used in determining the most adequacy of the fit of the models to the experimental data as shown in Table 2. Ana *et al* (2007) Method had satisfied two of the criteria with the most minimum value of *RMSE* (3.7713%) and highest *r* value (0.9933). However the *E* value of 11.52% which are higher than 10% requirement. This can be improves by further optimization of the model in the future.

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5. Conclusion

The research is conducted to find the suitable mathematical model in predicting the Cymbopogon *winterianus* essential oil extraction by steam distillation. The research is a continuation of Cassel and Vargas (2006) previous research "*Experiments and Modelling of the Cymbopogon winterianus Essential Oil Extraction by Steam Distillation*". The proposed optimum *D* parameter value in the previous research is used to simulate and verify the prediction value of the mathematical model. Laboratory scale experiment data in previous research done by Cassel and Vargas (2006) in their research "*Experiments and Modelling of the Cymbopogon winterianus Essential Oil Extraction by Steam Distillation*" were used as a benchmark and reference.

From the research, it is found that the mathematical model proposed by Cassel and Vargas (2006) does not have the perfect fit in predicting the citronella essential oil yield extracted over time. The RMSE error calculated for the mathematical model and parameter *D* value proposed is 0.317916 (31.79% error). By further optimizing the mathematical model, a new value of parameter *D* was proposed and the *RMSE* value reduces to 0.226 (22.6% error). Thus the objective to optimize the mathematical model minimum of 5% was achieved. Nonetheless the value of the RMSE error for the optimized mathematical model is still very high and more than 10% error in predicting the extraction process. This is due to the assumption the made earlier when developing the mathematical model where parameter *D* is assumed to be constant overtime and does not take into account the resistance occurs in diffusion of essential oil from the interior of the plant to the external surface of the plant.

Thus, in order to optimize the prediction of essential oil, four new mathematical models developed by Hervas *et al* (2006) Method, Garkal *et al* (2012) Method, Ana *et al* (2007) Method and Milojevi *et al* (2008) Method were studied. The adequacy of the fit of the models to the experimental data are analyzed by using three statistical criteria that are correlation coefficient (*r*) ,the root mean square error (*RMSE*) and the mean relative deviation modulus (E). The model fits the experimental data well when $RMSE \rightarrow 0$, $r \rightarrow 1$ and $E < 10\%$. Ana et al. (2007) Method had satisfied two of the criteria with the most minimum value of *RMSE* (3.7713%) and highest *r* value (0.9933). However the *E* value of 11.52% which are higher than 10% requirement. This can be improves by further optimization of the model in the future.

The analysis of the result from the optimization indicates that the mathematical model developed by Ana *et al* (2007) based on mass transfer fundamentals fitted very well the experimental data. The mathematical model is based on second order rate with two k parameter which each represents the washing and diffusion separately. The hypothesis used in developing the mathematical model is more coherent with the reality observed while the mathematical model developed by Cassel and Vargas (2006) based on the theory of diffusional model and Fick's Law in steady-state for one-dimensional rectangle geometry is more restrictive to describe the steam distillation process of extraction of Cymbopogon *winterianus* essential oil.

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