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# Kinetic Studies on Extraction of Essential Oil from Lemongrass Leaves (*Cymbopogon citratus*) by Steam Distillation Industrial Scale

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Study on kinetics and modeling of the essential oil steam distillation are required for the optimization of the energy requirement, parameters and the process scale-up. In this study, hydrodistillation for extraction of essential oils from lemongrass (*Cymbopogon citratus*) steam distillation was investigated. The recovery of essential oils was carried out on an industrial scale instrument and with with different material quantities. The experimental results are determined by the integral method of analysis. The kinetic parameters were evaluated from experimental data, which were generated at the different weights of the lemongrass. The extraction rate constant explaining the extraction efficiency achieved from this study is found to be $k_2$ of 0.0661 $min^{-1}$ in 710 kg using a second-order kinetic model. The GC/MS results revealed that 14 components were identified in *Cymbopogon citratusoils*. The oil is extremely rich in citral (69.775%), which obtained in 180 min.

**Keywords: Essential oil, Lemongrass leaves, *Cymbopogon citratus*, Steam distillation, Kinetic model, GC-MS analysis.**

## INTRODUCTION

Essential oils have become more prevalent in human life due to their benefit, creating a significant increase in demand for them worldwide. Essential oils are aromatic and volatile compounds, which are extracted from the seeds, flowers, leaves and stems [1-7]. Essential oils play a vital role in different fields including cosmetics, pharmaceuticals, food and beverage industry as they constitutes various natural properties such as antibacterial, antiviral, antifungal, insecticidal and anti-herbivore characteristics [8-14].

Lemongrass, also named as *Cymbopogon citratus* or *Andropogon citratus*, currently has about 55 species. The previous study reported that lemongrass essential oils play a crucial role in the treatment of different diseases such as oily skin, scabies, and acne due to their antibacterial and antimicrobial activities. Lemongrass could be used as a vegetable added in soup or as a medicinal herbto enhance the flavour and organoleptic properties [15-20]. The essential oil accounts for more than 75% of weight of the lemongrass. The component with the highest content is citral (which accounts for more than 80% of citronella oil content) which is very unstable and could be easily oxidized and denatured by external conditions such as light, heat and pH. Among essential oils isolated from various plant sources, lemongrass oil shows the highest antioxidant activity.

To obtain essential oils, there are different method of extraction such as supercritical fluid extraction, solvent extraction, hydrodistillation, and steam distillation. The hydrodistillation method has become increasingly common for extraction of essential oils from plant materials due to the simplicity of installations and ease of performing. Previous studies have demonstrated that the steam distillation is a suitable technique for extraction of essential oils from lavender and artemisia leaves. The hydrodistillation process has been being improved constantly through many studies dealing with issues of separation efficiency, composition and utility of essential oils. One type of such improvement studies is investigations on the kinetic model of distillation. Not only does a well-modelled kinetics of distillation control and improve extraction efficiency but it could also give a better insights on the process, from which adjustments on the existing system could be made [21-33].

In this study, a kinetic study conducted on a pilot distillation plant was carried out to examine the steam distillation process for the extraction of essential oil from lemongrass leaves (*Cymbopogon citratus*) growing Tien Giang Province, Vietnam.This study aimed to discover the kinetic model that best describes the distillation process of essential oils from the lemongrass leaves (*Cymbopogon citratus*). First-order model and second-order model equation were investigated and subsequently confirmed using experimental data.

## EXPERIMENTAL

**Extraction of essential oil:** Leaves of lemongrass, which is the by-product of the harvesting of commercial lemongrass, were collected in January, 2019 from Tien Giang province, Vietnam, at coordinates of 10º15′N 106º39′E. After harvest, lemongrass leaves were wilted to a moisture content of 50% ofthe original. At this moisture, lemongrass leaves were preserved for a certain days in a cool place.

**Steam distillation kinetics model:** In the study, kinetic modeling for extraction of lemongrass oil by steam distillation methods was performed using the first-order and second-order models (Table-1) [34-37].

**GC-MS analyses:** GC-MS is used to analyze the composition of the essential oils of all samples. About 25 µL of essential oil was mixed in 1.0 mL of *n*-hexane. GC Agilent 6890N, coupled with MS 5973 inert and HP5-MS column was the instrument in the study. Head column pressure was set to 9.3 psi. GC-MS system was performed hold under the following conditions: flow rate 1.0 mL/min; carrier gas He, split 1:100; injection volume 1.0 µL; injection temperature 250 ºC; 50 ºC for 2 min, then increased by 2 ºC/min to80 ºC, and increased by 5 ºC/min to 150 ºC, continue rising to 200 ºC at 10 ºC/min and rise to 300 ºC at 20 ºC/min for 5 min.

## RESULTS AND DISCUSSION

**Steam distillation of lemongrass essential oil:** Fig. 1 shows the cumulative oil yield results of the industry-scale hydrodistillation with respect to various experimental parameters. Overall, for fresh materials at varying weights from 639 to 710 kg, the oil yield curves obtained shared the similar trajectory where the diminishing oil yield is observed with increasing distillation time. The maximum oil yield observed from the curves approximated 0.273% (volume of attained essential oil/100 g raw materials used). Table-2 shows a summary of the maximum essential amount recovered from varying quantities of the materials (from 284 to 710 kg). More than 75% of essential oil was extracted within first 1.5 h each experiment. In the first experiment, materials were loosely packed in the distillation apparatus, while in all other experiments materials were tightly packed. All experiments were conducted with fresh materials without further cutting.The range of steam flow rate reported in each experiment is the range that allows efficient condensation with no excess steam. Steam distillation was carried out in each experiment for maximum period of 3 h and the oil yield is measured every 30 min. More than

TABLE-1
STEAM DISTILLATION KINETICS MODELS OF ESSENTIAL OILS FROM LEMONGRASS LEAVES

| Kinetics model | Kinetics equation | Linearized form of equation |
|---|---|---|
| First-order model | $\frac{dq_t}{dt} = k_1(q_o - q_t)$ | $\log(q_o - q_t) = \log(q_o) - \frac{k_1}{2.303}t$ |
| Second-order model | $\frac{dq_t}{dt} = k_1(q_o - q_t)^2$ | $\frac{t}{q_t} = \frac{t}{q_o} + \frac{1}{h}$ |

where $k_1$ ($min^{-1}$) is extraction the rate constant for first-order, $q_o$ (mL/100 g materials) is the lemongrass oil content initially present in the leaves, $q_t$ (mL/100 g materials) is the amount of essential oil extract until time t (min).

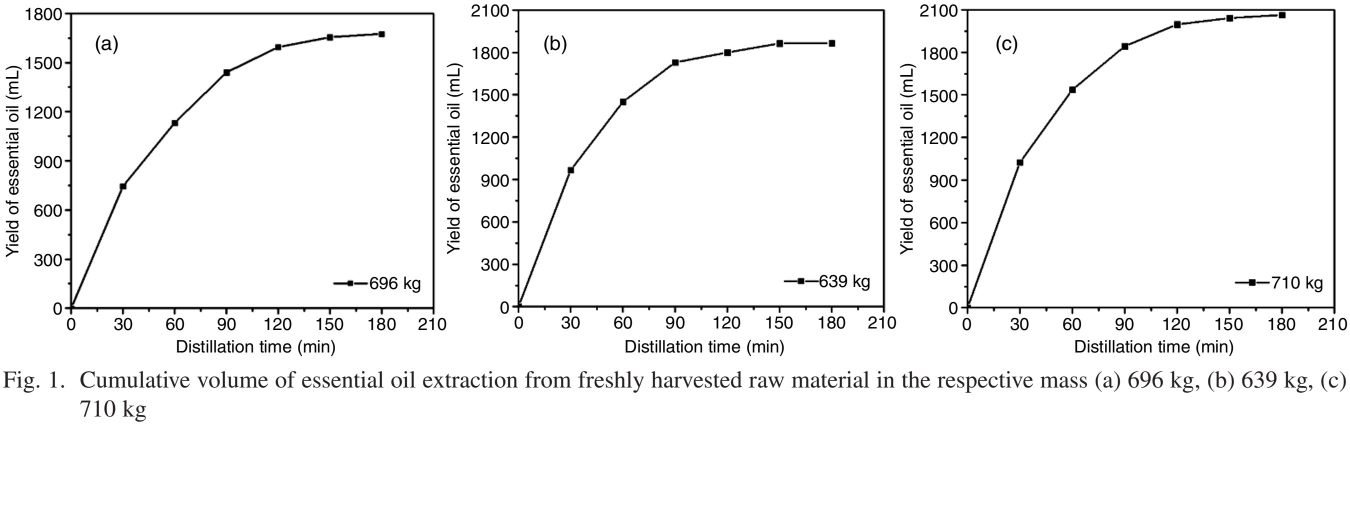


Fig. 1. Cumulative volume of essential oil extraction from freshly harvested raw material in the respective mass (a) 696 kg, (b) 639 kg, (c) 710 kg

TABLE-2
EXPERIMENTAL DATA FOR STEAM DISTILLATION OF LEMONGRASS OIL

| Exp. No. | Weight of material (kg) | Condition of material | Packing | Steam flow rate (L/h) | Oil produced after | | Oil yield (%) |
|---|---|---|---|---|---|---|---|
| | | | | | 1.5 h (mL) | 3 h (mL) | |
| 1 | 284 | Uncut | Loose | 105-115 | 720 | 760 | 0.267 |
| 2 | 696 | Uncut | Tight | 100-110 | 1440 | 1676 | 0.240 |
| 3 | 639 | Uncut | Tight | 110-130 | 1730 | 1867 | 0.292 |
| 4 | 710 | Uncut | Tight | 110-130 | 1843 | 2064 | 0.291 |

75% of essential oil was extracted within first 1.5 h each experiment.

When comparing the results from the first and the second experiment, it is evident that the oil yield from the second distillation attempt, where materials were packed, is lower. The reduced yield could be due to the fact that tightly packed materials both hinders steam contact with the materials and obstructs the up-ward oil-steam flow to the condensation system [38]. However, considering that this yield discrepancy is marginal and that compressing materials significantly reduces distillation time and costs and in turn enhance economic efficiency, the strategy of tight-packing of materials was selected in subsequent investigations. In 3$^{rd}$ and 4$^{th}$ experiments,we put in materials that were slightly more compressed than in the second experiment. In addition, we mechanically improved the cage of the material-containing tank to allow better passage and contact of the steam with the materials. The results showed that oil yields significantly improved with such adjustments.

**Modeling of the kinetic of essential oil distillation:** By plotting log ($q_o - q_t$) against t, the first-order kinetic model of the extraction process could be determined. Fig. 2 illustrates the essential oils of lemongrass leaves in a linear form of the first-order model. Visually, it is suggested from the plot that the extraction process could be linearized according to a first-order model. The linearization results of the first-order kinetic model for essential oils extraction along with $q_o$, $k_1$ and the coefficient of determination ($R^2$) were reported in Table-3. It was shown that $R^2$ of the first-order kinetic was relatively low, ranging 0.85-0.97 depending on the material mass. In addition, the fitting of the linear equation was apparently better at the early period of distillation suggested that the first-order is not suitable to describe the experimental data of this process. This is consistent with some other studies [39,40] in which inadequacy of the first-order model was articulated when it comes to describing processes having multiple mechanisms.

Since the distillation process may involve multiple mechanisms including wetting the materials and diffusion of oil within the materials to the solvent, the second-order kinetic model is applicable in this case. Similar to the previous investigation, plots of $t/C_t$ against t were established to determine the second-order kinetic on the extraction of lemongrass oil with the air flow rate 110-130 L/h (Fig. 3). In addition, the value $k_2$ and $q_o$ can also be derived by minimization of the sum of square of errors between the experimental data with model data. Table-4 shows parameters obtained from the mathematical model of the second-order kinetic model. The second-order kinetic model for lemongrass oil extraction by steam distillation method has relatively high coefficient of determination values, reaching

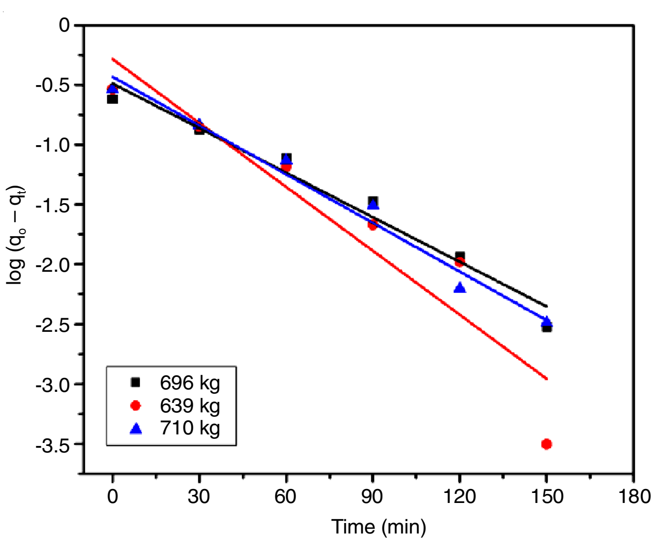

Fig. 2. First-order extraction kinetic of lemongrass

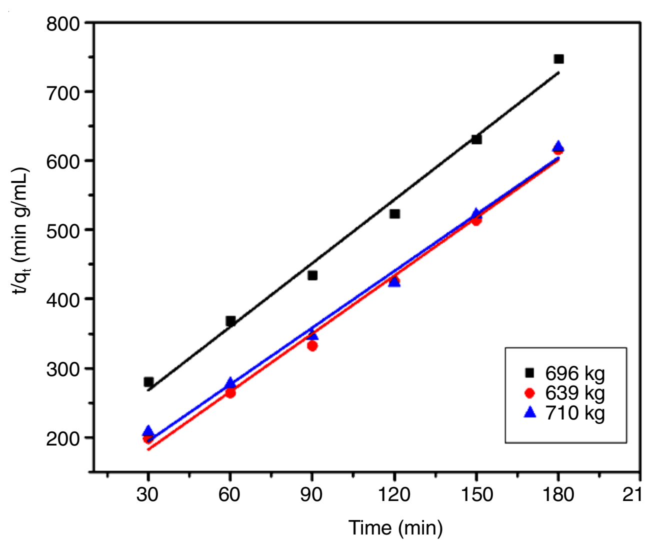

Fig. 3. Second-order extraction kinetics of lemongrass

TABLE-3
LINEARIZATION OF FIRST-ORDER KINETIC MODEL FORLEMONGRASS OIL EXTRACTION

| Mass (kg) | Calculation method | Slope* | $k_1$ (min$^{-1}$) | Intercept* | $q_0$ (mL/g) | $R^{2*}$ |
|---|---|---|---|---|---|---|
| 696 | Linear regression (Solver*) | -0.01243 | 0.0286 | -0.48838 | 0.3248 | 0.95939 |
| 639 | Linear regression (Solver*) | -0.01783 | 0.0411 | -0.28333 | 0.5208 | 0.85917 |
| 710 | Linear regression (Solver*) | -0.01358 | 0.0313 | -0.43205 | 0.3698 | 0.97156 |

*Values obtained from OriginPro 9.0

TABLE-4
LINEARIZATION OF SECOND-ORDER KINETIC MODEL FOR LEMONGRASS OIL EXTRACTION

| Mass (kg) | Calculation method | Slope* | $k_2$ ($min^{-1}$) | Intercept* | $q_0$ (mL/g) | $R^{2*}$ |
|---|---|---|---|---|---|---|
| 696 | Linear regression (Solver*) | 3.05853 | 0.0530 | 176.48733 | 0.3270 | 0.9888 |
| 639 | Linear regression (Solver*) | 2.79172 | 0.0787 | 98.711733 | 0.3582 | 0.99138 |
| 710 | Linear regression (Solver*) | 2.72975 | 0.0661 | 112.82267 | 0.3663 | 0.9913 |

*Values obtained from OriginPro 9.0

approximately 0.99, while this value of first-order model about 0.85-0.9. Therefore, the second-order kinetic model is more fitted to describe the experimental data of lemongrass oil extraction by steam distillation method.

**Kinetic steam distillation extraction by GC-MS:** The chemical components of the hydrodistilled essential oils from lemongrass were characterized by GC-MS. Six samples were analyzed at different intervals in the 4th experiment and the results are shown in Table-5. Overall, a total of 14 different compounds accounting more than 90% the oil content was identified in the essential oil. In the final obtained oil, the most representative components of lemongrass were β-myrcene (20.646%), β-citral (30.429%), α-citral (38.743%) and nerol (2.798%). Other significant compounds were β-ocimene (0.542%), β-linalool (0.855%), camphenehydrate (0.443%) and citronellol (0.345%). This is well-agreed with previous studies where neral and geranial were reported to be main constituents [41-43]. Previous studies also showed that α-citral and β-citral might modulate inflammatory processes and DNA damage and inhibited growth of various Gram-negative and Gram-positive bacteria [15,44].

Lemongrass oils obtained at all time intervals were consisted principally of β-myrcene, α-citral and β-citral, which primarily determine the oil quality and play an essential role in the antibacterial characteristic of the product. Interestingly, some compounds, such as compound 6, only present at longer times. Almost all minor compounds were identified in previous studies except for sulcatone. In addition, isoneral and isogeranial were the compounds that have not been previously detected. In addition, as observed in Fig. 4, the prolonging of distillation time seems to reduce the content of β-myrcene. However, the content of α-citral and β-citral was kept at reasonably high levels

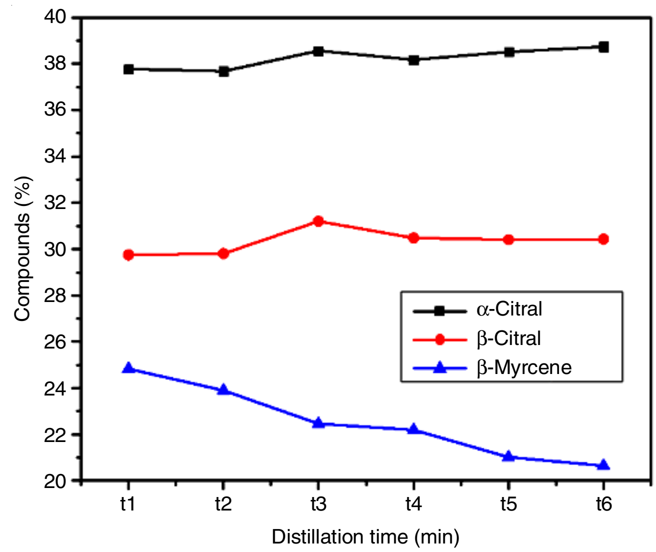


Fig. 4. Change of the main component of lemongrass oil

regardless of distillation time. From these results, an appropriate strategy to manipulate the content of the oil constituents could be devised.

## Conclusion

In this study, an industry-scale hydrodistillation of essential oils from Vietnamese lemongrass has been examined and kinetically modeled. Recovery of essential oils from lemongrass by steam distillation follows second order rate kinetics with an average rate constant $k_2$ of 0.0661 $min^{-1}$ with the material weight of 710 kg. This means that the level of oil in the lemongrass is directly proportional to the oil extracted per unit time. Fourteen components were identified in *Cymbopogon citratus*

TABLE-5
CHEMICAL COMPOSITION OF LEMONGRASS LEAVES OIL OBTAINED FOR 180 min AND AT DIFFERENT EXTRACTION TIME

| Retention time (min) | Compounds | Samples collected at different time | | | | | |
|---|---|---|---|---|---|---|---|
| | | 30 min (%) | 60 min (%) | 90 min (%) | 120 min (%) | 150 min (%) | 180 min (%) |
| 9.949 | Sulcatone | 0.897 | 1.021 | 0.852 | 1.031 | 1.185 | 1.192 |
| 10.126 | β-Myrcene* | 24.830 | 23.893 | 22.46 | 22.191 | 21.014 | 20.646 |
| 12.636 | β-Ocimene* | 0.472 | 0.548 | 0.395 | 0.532 | 0.551 | 0.542 |
| 16.317 | β-Linalool* | 0.883 | 0.824 | 0.758 | 0.758 | 0.829 | 0.855 |
| 19.046 | Camphenehydrate* | 0.455 | 0.481 | 0.404 | 0.484 | 0.574 | 0.443 |
| 19.391 | Not identified | tr | 0.280 | 0.245 | 0.291 | 0.301 | 0.324 |
| 19.977 | Isoneral | 1.026 | 1.228 | 1.109 | 1.165 | 1.371 | 1.392 |
| 20.834 | Isogeranial | 1.33 | 1.650 | 1.373 | 1.744 | 1.825 | 1.877 |
| 22.863 | Citronellol* | 0.182 | 0.247 | 0.204 | 0.31 | 0.334 | 0.345 |
| 23.313 | β-Citral* | 29.754 | 29.807 | 31.206 | 30.484 | 30.408 | 30.429 |
| 23.856 | Nerol* | 1.901 | 2.071 | 2.131 | 2.455 | 2.694 | 2.798 |
| 24.473 | α-Citral* | 37.782 | 37.686 | 38.569 | 38.182 | 38.518 | 38.743 |
| 29.137 | Caryophyllene* | 0.107 | 0.132 | 0.137 | 0.197 | 0.201 | 0.208 |
| 29.628 | α-Bergamotene* | 0.114 | 0.131 | 0.154 | 0.175 | 0.194 | 0.207 |

*Component identified by other authors [8-11] oils. The major components were citral (69.775%) which was found in the oil sample at 180 min of distillation time. In addition, it was found that the distillation time greatly curbs the content of β-myrcene in the obtained oil.

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## CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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