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EFFECT OF MATERIAL LENGTH ON KINETICS OF ESSENTIAL OIL HYDRODISTILLATION FROM LEMONGRASS (*Cymbopogon citratus*)

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Abstract

The objective of this study is to investigate the effect of material length on the kinetics of the hydrodistillation of lemongrass (*Cymbopogon citratus*) essential oil with the use of a laboratory-scale hydrodistillation unit. The experimental results showed that the highest yield was taking place at a material length of 10 cm. Based on simultaneous evaporation and diffusion mechanism, the kinetics of lemongrass oil hydrodistillation process can be described mathematically using a two-parameter model. It was found that the parameter relating to the fast oil distillation period (evaporation coefficient) was larger than that of the slow oil distillation period (diffusion coefficient), thus, the evaporation stage was much faster than the diffusion stage. The gas chromatography-mass spectrometry analyses have proved that the material length did not influence the composition of lemongrass oils. The content of the citral as the main component in lemongrass essential oils was found to be more than 80%.

Keywords: Essential oil, Hydrodistillation, Kinetic, Lemongrass.

1. Introduction

Lemongrass (*Cymbopogon citratus*) is a tropical plant found in South and Southeast Asia. It is cultivated on a large scale as a source of essential oil, especially in the tropics and subtropics, around the world. The leaves are used in traditional medicine and are often found in herbal supplements and teas.

Lemongrass is a plant of the grass family that contains about 1 to 2% essential oil on a dry weight basis. The lemongrass essential oil is characterised by a high content of citral, which is a natural combination of isomers neral and geranial, two isomeric aldehydes. The lemongrass essential oil has a number of uses including for vitamin A, perfumes, insect sprays, cosmetics, food and drinks. It is very important in the perfumery industry because it blends well with a great variety of essential oils [1] and it is also widely used as an additive in a variety of consumer products [2]. Lemongrass oil has antifungal properties and can, therefore, be used as a preservative for food. Recently, Supardan et al. [3] reported that a cassava starch edible film containing lemongrass oil may extend the shelf life of food.

The lemongrass oil can be produced from fresh or dried plant material using various methods of extraction including pressurised liquid extraction using hexane [1], hydrodistillation [4-6], solvent extraction using supercritical fluid carbon dioxide [7] and high-pressure carbon dioxide extraction [8]. Distillation using water or steam is a traditional method for extraction of essential oils [9], although this method is energy intensive and suffers from long extraction times and the loss of volatile components. However, distillation using water or steam is economically desirable because it does not involve solvents, thereby avoiding solvent evaporation. This method is also simple and safe to operate, environmentally friendly [9] and can be employed by both small and large industries [10, 11].

In order to determine a more efficient process, it is necessary to develop a new method, which can speed up the process of essential oil extraction with a higher yield whilst maintaining good quality. Some new essential oil extraction methods have been developed, including microwave hydrodistillation and microwave steam distillation [12, 13]. A high-performance extraction technique is required to achieve a high oil yield due to the relatively low concentration of lemongrass oil in the plant, with the yield depending on the conditions under which, the extraction takes place, such as the time scale of the process as well as the size and the drying method of the material [14]. In addition, many variables affect the essential oil's chemical composition, for example drying procedure, a method of extraction, storage conditions and the analysis conditions that are used for the identification of the compounds [15].

There is little information in the published literature on the kinetics of lemongrass oil hydrodistillation. This might be due to the fact that producers of lemongrass oil use conventional technologies, with only a little concern for process optimisation. Knowledge of the kinetic model of lemongrass oil hydrodistillation is however, very important for the technological and economic analysis of the process. Such knowledge would contribute to the fundamental understanding of the process and may lead to better process control and higher efficiency of the process [16]. Many researchers have modelled the kinetics of essential oil hydrodistillation from plant materials, such as flowers of *Lavandula officinalis* L. [15], *Juniperus communis* L. [16] and *Pistacia lentiscus* L. [17].

The aim of this study is to investigate the kinetics of lemongrass essential oil hydrodistillation, with the main goal being to study how the material length affects the yield and kinetics of the hydrodistillation process. The kinetic parameters can be used to predict the amount of essential oil extracted and a study of the kinetics was a necessary first step before developing and implementing a complete industrial process.

2. Experimental

2.1. Materials

Lemongrass (*Cymbopogon citratus*) plants were collected from a local supplier in Banda Aceh, Indonesia then air was dried. After drying, the stems and leaves of the lemongrass were cut into small pieces with a specified length and then stored at room temperature until required. The anhydrous sodium sulphate used in the experimental work was of analytical grade from Merck and used as received. Distilled water was used in this study. The lemongrass was soaked in distilled water for 20 min before the extraction performed to improve the collection efficiency.

2.2. Experimental procedure

The experimental procedure for lemongrass oil hydrodistillation is presented in Fig. 1. The experiments were conducted in a laboratory-scale hydrodistillation unit containing a grid that keeps the plant material above the boiling distilled water below. The wet steam produced by the water passes through the plant material. The still was packed evenly and not too tightly so that the steam efficiently extracts the essential oil from the plant material.

The hydrodistillation process was performed at a temperature of 100 ^{o}C for 165 min. To study the effect of material length on the amount of essential oil produced, the hydrodistillation process was carried out at three material lengths, namely 5, 10 and 15 cm. Each sample was treated in the same way. In each experiment, the still was filled with about 700 g of lemongrass. Successive withdrawals of essential oil fractions were made at different times. The mass of oil obtained was recorded. During the process, steam and volatile components rise from the still to the condenser, in which, two immiscible liquid phases of aromatic water and lemongrass oil are formed. The aromatic water and lemongrass oil were separated using a separating funnel. Then, the lemongrass essential oil obtained was dried over anhydrous sodium sulphate to remove any remaining water, weighed and stored in a sealed vial at 4 °C for further analyses.

The yield of lemongrass essential oil was determined as the mass of essential oil produced (g) per dry raw material in the hydrodistillation still, as presented by Eq. (1). Meanwhile, oil content initially present in the material, q_0 (g/g) was determined using a Clevenger-type apparatus. The system was operated at a temperature of 100 ^{o}C, the power of 500 W for about 5 h under atmospheric pressure. The experiments were repeated twice to ensure data reproducibility, and the average value for the measured data was presented. The experimental error found to be less than 5% for the whole experiments, thus, the experimental error among the collected data at the same condition was negligible.

$$\text{Essential oil yield} = \frac{\text{Weight of oil}}{\text{Weight of dry raw material}} \times 100\% \quad (1)$$

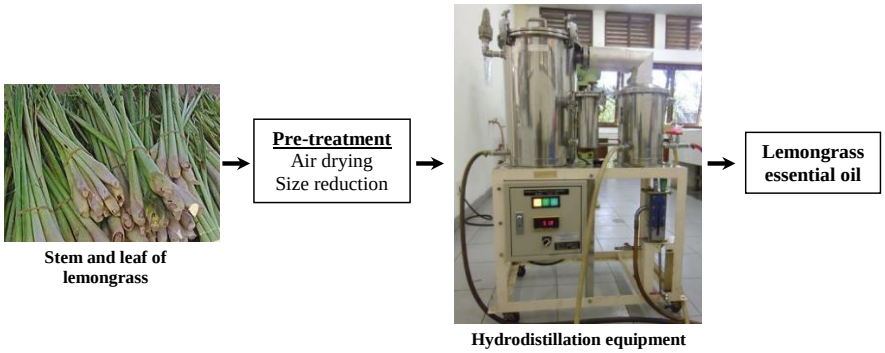

Fig. 1. Experimental procedure of lemongrass oil hydrodistillation.

2.3. **Product analysis**

The composition of the lemongrass essential oil was analysed using gas chromatography-mass spectrometry (GC-MS) (Model QP 5090A, Shimadzu, Japan), with a capillary column DB5 (30 m, 0.25 mm, 25 μm). The carrier gas was helium. The compounds were characterized by database matching and comparison of their MS spectra with an existing database (Wiley and National Institute of Standards and Technology libraries). The composition of the lemongrass oil was determined using the normalization method without a correction factor. Then, the percentage of peak area for each compound as a function of the total peak area from all compounds was determined and reported as the relative amount of that compound.

3. Results and Discussion

3.1. Yield of lemongrass oil

The lemongrass oil obtained from each experiment was very similar in appearance; dark yellow in colour with a characteristic lemony flavour and a watery viscosity. The distillation time had an important influence on the yield of essential oil from the lemongrass, with Fig. 2 shows the effect of distillation time on the essential oil yield for various material lengths. For the given conditions, there was a marked correlation between the distillation time, material length and essential oil yield as in Fig. 2. The yield of lemongrass essential oil increased with increasing distillation time, which can be explained by the fact that an increase in the distillation time provides to a longer contact between the steam and the plant material, thereby increasing mass transfer [14].

As can be seen in Fig. 2, after 120 minutes of distillation, there was no longer any increase of the essential oil yield with a further increase in distillation time. It can be concluded that when the material was extracted for 120 minutes, the remaining essential oil contained in the lemongrass had dropped to a minimum value. Similar trends have been found by Sukardi et al. [18] in regard to the extraction of patchouli oil and Hasmita et al. [14] for extraction of ginger oil.

Figure 2 also presents the effect of material length on the yield of lemongrass essential oil. There was no significant difference in essential oil yield at 30 min of distillation time. As the distillation time increased, however, the effect of the material

length on the yield of essential oil became more pronounced. As expected, the yield increased with decreasing material length from 15 to 10 cm. This can be explained by the fact that decreasing the size of the materials leads to a higher surface area, making extraction more efficient. Kusuma and Mahfud [19] reported a similar trend for the extraction of essential oil from *Pogostemon cablin* Benth. In our experiments, however, the yield decreased with a further decrease of the material length to 5 cm. It is supposed that a size reduction process would not be suitable for the efficient extraction of lemongrass essential oil because a part of the volatile compounds will be lost from the plant material during the size reduction pre-treatment process. In addition, the possibility of overall bed compaction for small material size will reduce contact between the steam and the plant material, thereby decreasing mass transfer. Langa et al. [20] also reported a similar trend for the production of Spanish sage essential oil by supercritical fluid extraction technique.

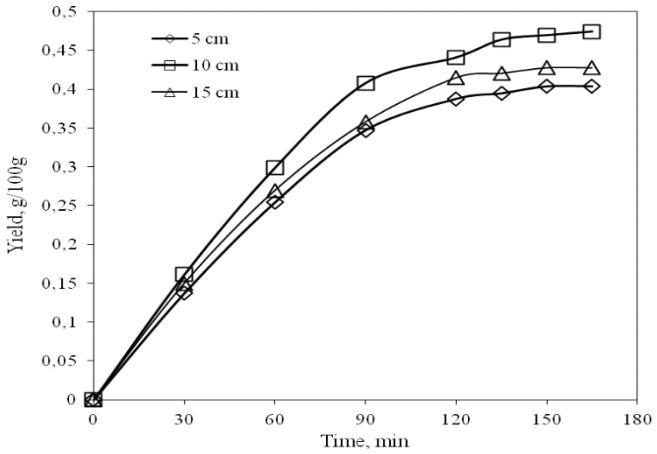

Fig. 2. Effect of distillation time on essential oil yield for various material lengths.

3.2. Hydrodistillation kinetics

The time course of the change in the lemongrass oil yield during the distillation process for different material lengths is shown in Fig. 2 and is a typical curve for the distillation of essential oils from an aromatic plant. As can be seen in Fig. 2, two periods of lemongrass oil distillation can be readily observed, i.e., fast oil and slow oil distillation. The fast oil distillation period occurs during the initial stage of distillation and during this period it is assumed that the essential oil was rapidly liberated and evaporated from the external surfaces of the plant materials. In this period, the yield of lemongrass oil increased rapidly. The rate of distillation slowed down until a nearly constant oil yield was reached with the progress of the distillation process. In the latter period, slow molecular diffusion of the essential oil from the undestroyed cells to the surfaces of the plant materials was followed by oil distillation. The rate of oil distillation slowed because the diffusion rate of essential oil was slow. This mechanism was important for subsequent modelling of the extraction kinetics. Some studies also reported a similar mechanism, such as Hasmita et al. [14] in the extraction of essential oil from ginger and Mebrouk et al. [17] in the extraction of essential oil from Algerian *Pistacia lentiscus* L.

Various models have been developed to describe the solid-liquid extraction process [21]. Physical kinetic models are based on the mass transfer phenomenon through the solid plant materials and from their surfaces into the bulk of the solvent. The film theory and unsteady diffusion through the plant material are the most frequently used kinetic models.

Milojevic et al. [16] further developed a mathematical model representing the kinetics of essential oil hydrodistillation. This model was based on a solvent extraction process in which, the evaporation and diffusion stages took place simultaneously. The model, defined by Eq. (2), involves two parameters, where *b* represents the stage of fast oil distillation (evaporation coefficient), and *k* represents the stage of slow oil distillation (diffusion coefficient). This model is the best choice for representing the essential oil hydrodistillation kinetics of any type of plant material.

$$\frac{q_0 - q}{q_0} = (1-b)\cdot e^{-kt} \tag{2}$$

The linearized form of the Eq. (2) can be utilised to determine the two parameters *b* and *k* in Eq. (3).

$$\ln\left(\frac{q_0 - q}{q_0}\right) = \ln(1-b) - kt \tag{3}$$

By plotting $\ln\left(\frac{q_0 - q}{q_0}\right)$ versus time (*t*), one can obtain –*k* as the slope and ln (1-*b*) as the intercept with the *y*-axis.

As can be seen in Fig. 3, the linearized form of all of the curves corresponding to the different material lengths verifies the kinetic model. The performance of the kinetic model was determined based on the value of the coefficient of linear correlation (R^2) [22].

The kinetic model has a relatively high coefficient of linear correlation (> 0.9), from which, it can be concluded that Eq. (3) represents quite well the experimental data of the hydrodistillation of lemongrass essential oil. Then, the kinetic model parameters, *b* and *k*, were obtained from the slope and intercept of each curve. The results are presented in Table 1.

Both kinetic parameters for the 10 cm material length were larger than for the 5 cm and 15 cm material lengths. Based on the kinetic parameters, it can be concluded that the best condition was found at a material length of 10 cm.

It was also observed that the *k* value was smaller than *b* for all material lengths; thus, it can be concluded that the rate of the process was controlled by the slow oil distillation period. The coefficient of the fast oil distillation period was 38 to 42 times larger than the coefficient of the slow oil distillation period, indicating that the diffusion was much slower than the evaporation.

Table 1. Values of kinetic parameter.

Material length (cm)	*b* (-)	*k* (1/min)
5	0.1139	0.0027
10	0.1293	0.0034
15	0.1189	0.0030

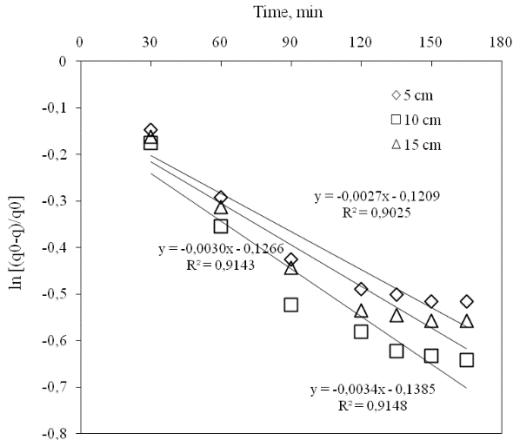

Fig. 3. Independence of $\ln\left(\frac{q_0 - q}{q_0}\right)$ on time.

3.3. Composition of lemongrass oil

From the GC-MS analyses, 16 compounds were identified in the lemongrass oils. In the experimental range, however, the lemongrass essential oil composition showed no significant differences for the different material lengths. As presented in Table 2, there were 7 components with a concentration of higher than 1%. Independently of their origin, lemongrass essential oils contain from 30 to 93.74% citral, with geranial being the main component [23].

Citral is a mixture of the bioactive isomers neral and geranial. GC-MS analysis results showed that the content of the citral compound (neral and geranial) in the lemongrass essential oils obtained by hydrodistillation was more than 80%. Citral, limonene, citronellal, myrcene and geraniol have been reported as marker compounds in lemongrass essential oils. The marker compounds are components of the chemical compounds responsible for the biological activity [24] and can be used as references for quality control evaluation. Experimental results showed that neither limonene nor citronellal was found in the analysed samples. Barbosa et al. [23] also observed an absence of citronellal and limonene in Brazilian lemongrass essential oils. The chemical composition of lemongrass oil is influenced by several factors, such as genetic diversity, habitat, climatic, seasonal as well as experimental conditions applied [5, 8].

Table 2. Effect of material length on composition of lemongrass oil.

Component	Composition (%)		
	Material length 5 cm	Material length 10 cm	Material length 15 cm
Myrcene	10.86	10.84	10.77
3,4-Octadiene, 7-methyl	1.0	1.3	1.42
4,5-Nonadiene, 2-methyl	1.64	1.94	2.01
Neral	32.05	32.69	31.87
Geraniol	1.16	1.45	1.07
Geranial	50.55	49.61	49.08
Geranil asetat	1.37	2.17	2.22

4. Conclusions

It was observed that the process variables of distillation time and material length have a remarkable effect on the yield of lemongrass oil. The yield of lemongrass essential oil increased with increasing distillation time. The lemongrass should be extracted with the material length of 10 cm to achieve an optimum mass transfer and highest yield. The mechanism for lemongrass essential oil hydrodistillation is confirmed to occur in two steps, namely the evaporation of the oil from the surface and diffusion of oil from the internal parts of the plant material. A kinetic model using two parameters is used to describe the lemongrass oil hydrodistillation process. In the range of experimental studied, the parameter of the fast oil distillation period was 38 to 42 times larger than the parameter of the slow oil distillation period, indicating that the diffusion was much slower than the evaporation. It can be concluded that the rate of the process was controlled by the slow oil distillation period. GC–MS analyses of the lemongrass essential oils led to the identification of 16 compounds, with citral as the main component, accounting for more than 80% of the content. The experimental results showed that the composition of lemongrass essential oil was virtually the same for different lengths of material.

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Nomenclatures

b	Evaporation coefficient (-)
k	Diffusion coefficient (1/min)
q	Oil yield at time t (g/g)
q_0	Oil yield at time t (g/g)
R^2	Coefficient of linear correlation (-)

Abbreviations

GC-MS	Gas chromatography-mass spectrometry

References

1. Nur Ain, A.H.; Haiyee, Z.A.; Zahrah, M.S.H.; and Saim, N. (2013). An experimental design approach for the extraction of lemongrass (Cymbopogon citratus) oleoresin using pressurised liquid extraction (PLE). *International Food Research Journal*, 20(1), 451-455.
2. Lalko, J.; and Api, A.M. (2008). Citral: Identifying a threshold for induction of dermal sensitization. *Regulatory Toxicology and Pharmacology*, 52(1), 62-73.
3. Supardan, M.D.; Annisa, Y.; Arpi, N.; Satriana; and Mustapha, W.A.W. (2016). Cassava starch edible film incorporated with lemongrass oil: Characteristics and application. *International Journal on Advanced Science, Engineering and Information Technology*, 6(2), 216-220.
4. Moncada, J.; Tamayo, J.A.; and Cardona, C.A. (2014). Techno-economic and environmental assessment of essential oil extraction from Citronella

*(*Cymbopogon winteriana*)* and Lemongrass *(*Cymbopogon citrus*):* A Colombian case to evaluate different extraction technologies. *Industrial Crops and Products*, 54, 175-184.

5. Mirghani, M.E.S.; Liyana, Y.; and Jamal, P. (2012). Bioactivity analysis of lemongrass (Cymbopogan citratus) essential oil. *International Food Research Journal*, 19(2), 569-575.
6. Hanaa, A.R.M.; Sallam, Y.I.; El-Leithy, A.S.; and Aly, S.E. (2012). Lemongrass (Cymbopogon citratus) essential oil as affected by drying methods. *Annals of Agricultural Sciences*, 57(2), 113-116.
7. Carlson, L.H.C.; Machado, R.A.F. Spricigo, C.B.; Pereira, L.K.; and Bolzan, A. (2001). Extraction of lemongrass essential oil with dense carbon dioxide. *Journal of Supercritical Fluids*, 21(1), 33-39.
8. Paviani, L.; Pergher, S.B.C.; and Dariva, C. (2006). Application of molecular sieve in the fractionation of lemongrass oil from high-pressure carbon dioxide extraction. *Brazilian Journal* of *Chemical Engineering*, 23(2), 219-225.
9. Guenther, E. (1948). *The essential oils* (Vol. 1). New York, United States of America: D. Van Nostrand Company, Inc.
10. Liu, J.; Wu, J.; Zhao, Y.X.; Deng, Y.Y.; Mei, W.L.; and Dai, H.F. (2008). A new cytotoxic 2-(2-phenylethyl) chromone from Chinese eaglewood. *Chinese Chemical Letters*, 19(8), 934-936.
11. Manzan, A.C.C.M.; Toniolo, F.S.; Bredow, E.; and Povh, N.P. (2003). Extraction of essential oil and pigments from Curcuma longa L. by steam distillation and extraction with volatile solvents. *Journal of Agricultural and Food Chemistry*, 51(23), 6802-6807.
12. Stashenko, E.E.; Jaramillo, B.E.; and Martinez, J.R. (2004). Comparison of different extraction methods for the analysis of volatile secondary metabolites of Lippia Alba (Mill.) N.E. Brown, grown in Colombia, and evaluation of its in vitro antioxidant activity. *Journal of Chromatography A*, 1025(1), 93-103.
13. Sahraoui, N.; Vian, M.A.; Bornard, I.; Boutekedjiret, C.; and Chemat, F. (2008). Improved microwave steam distillation apparatus for isolation of essential oils: Comparison with conventional steam distillation. *Journal of Chromatography A*, 1210(2), 229-233.
14. Hasmita, I.; Adisalamun, A.; Alam, P.N.; Satriana, S.; Mahlinda, M.; and Supardan, M.D. **(**2015**).** Effect of drying and hydrodistillation time on the amount of ginger essential oil. *International Journal on Advanced Science, Engineering and Information Technology*, 5(5), 300-303.
15. Stanojevic, L.; Stankovic, M.; Cakic, M.; Nikolic,V.; Nikolic, L.; Ilic, D.; and Radulovic, N. (2011). The effect of hydrodistillation techniques on yield, kinetics, composition and antimicrobial activity of essential oils from flowers of *Lavandula officinalis* L. *Hemijska Industrija*, 65(4), 455-463.
16. Milojevic, S.Z.; Stojanovic, T.D.; Palic, R.; Lazic, M.L.; and Veljkovic, V.B. (2008). Kinetics of distillation of essential oil from comminuted ripe juniper (Juniperus communis L.) berries. *Biochemical Engineering Journal*, 39(3), 547-553.
17. Mebrouk, K.; Saibi, S.; Nacer-Bey, N.; and Benyoussef, E.-H. (2012). Modelling of water distillation kinetic of Pistacia lentiscus L. essential oil. *Journal of Essential Oil Bearing Plants*, 15(6), 980-987.

18. Sukardi; Soeparman, S.; Argo, B.D., Irawan, Y.S. (2017). Optimization of patchouli oil (Pogostemon cablin Benth) with steam distillation assisted by pulsed electric field via response surface methodology. *Journal of Engineering Science and Technology* (JESTEC), 12(8), 2106-2119.
19. Kusuma, H.S.; and Mahfud, M. (2017). Microwave hydrodistillation for extraction of essential oil from Pogostemon cablin Benth: Analysis and modelling of extraction kinetics. *Journal of Applied Research on Medicinal and Aromatic Plants*, 4, 46-54.
20. Langa, E.; Porta, G.D.; Palavra, A.M.F.; Urieta, J.S.; and Mainar, A.M. (2009). Supercritical fluid extraction of Spanish sage essential oil: Optimization of the process parameters and modelling. *The Journal of Supercritical Fluids*, 49(2), 174-181.
21. Hamzah, M.H.; Man, H.C.; Jamaludin, H.; and Abidin, Z.Z. (2012). Optimization and kinetics of essential oil extraction from citronella grass by ohmic heated hydro distillation. *International Journal* of *Chemical Engineering and Applications*, 3(3), 173-177.
22. Baghdadi, Y.M.; and Hii, C.L. (2017). Mass transfer kinetics and effective diffusivities during cocoa roasting. *Journal of Engineering Science and Technology* (JESTEC), 12(1), 127-137.
23. Barbosa, L.C.A.; Pereira, U.A.; Martinazzo, A.P.; Maltha, C.R.A.; Teixeira, R.R.; and Melo, E.D.C. (2008). Evaluation of the chemical composition of Brazilian commercial Cymbopogon citratus (D.C.) stapf samples. *Molecules*, 13(8), 1864-1874.
24. Schaneberg B.T.; and Khan, I.A. (2002). Comparison of extraction methods for marker compounds in the essential oil of Lemon Grass by GC. *Journal of Agricultural and Food Chemistry*, 50(6), 1345-1349.