

Comparison of Citronella Oil Extraction Methods from *Cymbopogon nardus* Grass by Ohmic-heated Hydro-distillation, Hydro-Distillation, and Steam Distillation

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A proposed method for citronella oil extraction was developed with the application of ohmic heated hydro-distillation. The objective was to compare the performance of three different extraction methods, viz. ohmic heated hydro-distillation, hydro-distillation, and steam distillation. The maximum amount of extracted oil yield by ohmic heated hydro-distillation was 7.64 mL/kWh as compared to hydro-distillation and steam distillation methods that resulted oil yields of 3.87 mL/kWh and 1.69 mL/kWh, respectively. The kinetics of extraction followed a second-order model. Gas chromatography-mass spectrometry analysis found that the major constituents of citronella oil (GC-MS) for the different extraction methods were citronellal, citronellol, and geraniol. Scanning electron microscopy (SEM) of citronella grass provided evidence that the lignocellulosic sources of the extracted citronella oil were schizogenous cavities and cellular lignin. The citronella that had undergone ohmic-heated hydro-distillation and steam distillation showed some microfractures and less cell wall degradation than hydro-distillation. The cell walls were less rigid using ohmic-heated hydro-distillation compared to steam distillation. However, the cell walls of the hydro-distillation sample were less dense and exhibited pronounced swelling, but did not show any microfractures.

Keywords: Kinetics; Ohmic-heated hydro-distillation; Extraction; Citronella structural changes

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INTRODUCTION

Cymbopogon nardus, also known as serai wangi or citronella grass, is native to South India and Sri Lanka. It can also be found growing wild in most tropical Asian countries, Central America, and Africa. Recently, this plant has been planted commercially by entrepreneurs for essential oil extraction (Abdul Rahman Azmil *et al.* 2005). Citronella oil is considered one of the 20 most important essential oils that are traded globally (Lawrence 1993). Nowadays, world production of citronella oil is approximately 5000 tonnes, the bulk of which is produced in Taiwan, Guatemala, Malaysia, Brazil, Ceylon, India, Argentina, Ecuador, Madagascar, Mexico, and the West Indies (Skaria *et al.* 2007). Due to the market potential of citronella oil, its production can realise rural-sector participation in the citronella oil and aromatherapy industry (Coronel *et al.* 1984; Torres and Tio 2001). On the other hand, the citronella oil has been in

comparatively high demand because of its wide usage in the perfume, soap manufacturing, cosmetics, and flavouring industries, and is effective as an insect repellent (Duke and du Cellier 1993; Pandey 2010; Silva *et al.* 2011).

In addition, citronella grass is a non-woody plant and has lignocellulosic fiber characteristics (Kaur *et al.* 2010). The cell wall is made of three major lignocellulosic constituents: cellulose, hemicelluloses, and lignin (Wan Aizan *et al.* 2008). The cell wall is low in pectin and glycoproteins (Moerschbacher and Mendgen 2001). Citronella is classified as a monocotyledon and has a less complex system of fibers and associated botanical components than dicotyledons. Furthermore, less lignin is found in this lignocellulosic grass than in wood (Kaur and Dutt (2013).

Hydro-distillation and steam distillation are the most common or conventional methods for extracting oil from citronella (Brugnera *et al.* 2003; Abena *et al.* 2007; Mahalwal and Ali 2003; Nakahara *et al.* 2003; Cassel and Vargas 2006; Chungsamarnyart and Jiwajinda 1992). Although the process is very simple, it can induce thermal degradation, hydrolysis, and water solubility of some fragrance constituents (Reverchon and Marco 2006). Supercritical fluid extraction has also been introduced to extract citronella essential oil (Silva *et al.* 2011). However, this method has drawbacks such as high investment costs and a new and unfamiliar method of operation (Mohamed and Mansoori 2002). Additionally, due to its low polarity, the extraction of polar analytes is difficult. This limitation may be overcome by adding small amounts of polar modifiers, such as methanol or ethanol, to the supercritical fluid. These measures enhance the solubility of solutes in supercritical fluid extraction, thus increasing the extraction efficiency (Xiao *et al.* 2007).

Due to the success of natural herbs and sales of environmentally friendly products with encouraging returns, many entrepreneurs and herbal companies are currently developing plantations of aromatic plants such as citronella grass (Nor Azah 2000). The commercialisation of citronella oil has led to the formation of small-scale citronella oil extraction facilities (Torres and Tio 2001). Ohmic-heated hydro-distillation has been proposed as an alternative method for citronella oil extraction because the proposed prototype size is suitable for small- or medium-scale extraction facilities. Ohmic-heated hydro-distillation has been reported as an environmentally friendly extraction or “green” method because it does not necessitate the use of any organic solvent for extraction and requires substantially less energy to operate (Gahavian *et al.* 2012).

The objectives of this study were to propose and verify the kinetic models and performance of three extraction methods, namely, ohmic-heated hydro-distillation, hydro-distillation, and steam distillation, in terms of the oil yield and the chemical, physical, and scanning electron microscopy properties of citronella.

EXPERIMENTAL

Citronella Grass Preparation

Citronella grass from Ceylon (*Cymbopogon nardus*) was obtained from the Malaysian Agricultural Research and Development Institute (MARDI), Kuala Linggi, Kuala Sungai Baru, Melaka. The citronella grass was cut approximately 15 cm from the plant base or ground level. Before extraction, 4 kg of citronella grass was crushed by a EUMA™ Three-Phase Asynchronous Motor Y90L-2 crusher. The crushed citronella grass was immediately put into the distiller unit.

Kinetics Study

For the first hour of extraction, the extracted oil was determined at 15-min intervals. During the following hour, the extracted oil was determined at 30-min intervals until it reached equilibrium. At the end of each interval, the extracted oil yield was estimated as follows,

$$\text{Extracted oil yield (mL/kg)} = V_0/M_S \quad (1)$$

where V_0 is the volume of citronella oil extracted from the sample (mL) and M_S is the mass of the sample (kg).

Set-up of Ohmic-heated Hydro-distillation Unit

The ohmic-heated hydro-distillation system is shown in Fig. 1. The ohmic-heated hydro-distiller unit, which was 1.35 m long and had a capacity of 30 L, consisted of a central stainless steel 316 electrode (Fig. 2) that was 0.73 m long and 0.02 m in diameter in the middle of the distiller. An electrically insulated outer shell served as the outer electrode. The electrodes were connected to a three-phase alternating current step-down transformer 415/133/87-V power supply rated at 10 kVA with a maximum current of 60 A. Within the maximum current, two types of voltages could be used, *i.e.*, 77 V and 50 V. The distillation unit was connected to a multi-tube condenser made from stainless steel 304. The extraction parameters used were a power input of 77 V up to the boiling point of water, followed by 50 V until the end of the extraction, a water-to-citronella ratio of 3:1, and a crushing frequency of once.



Fig. 1. The ohmic-heated hydro-distillation system: (1) ohmic-heated hydro distiller unit, (2) power supply, (3) multi-tube condenser unit, (4) temperature sensor, (5) clamp meter

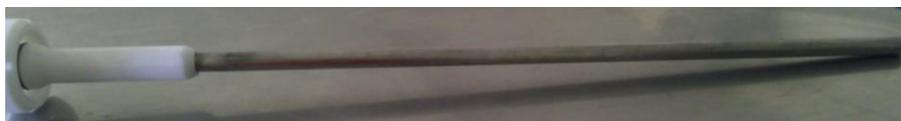


Fig. 2. A central stainless steel 316 electrode

Hydro-distillation

Hydro-distillation was carried out using a similar distillation unit to the one shown above, except that the central electrode was replaced with a 3kW/3-phase heating element (Fig. 3). The extraction parameters were a water-to-citronella ratio of 3:1 and a crushing frequency of once.



Fig. 3. A 3kW/3 phase heating element

Steam Distillation

The extraction was performed using the same distillation unit as in Fig. 1 except that the central electrode was taken out from that unit and replaced with a perforated grid plate (Fig. 4(a)). The perforated grid plate was placed at the bottom of the distillation unit, and the citronella grass was placed on the grid. Saturated steam generated from the boiler was injected through the perforated grid plate, separating the steam inlet at the bottom of the distillation unit (Fig. 4(b)). The steam in the distillation unit was at atmospheric pressure, and the temperature was 100 °C. A single crushing motion was used.

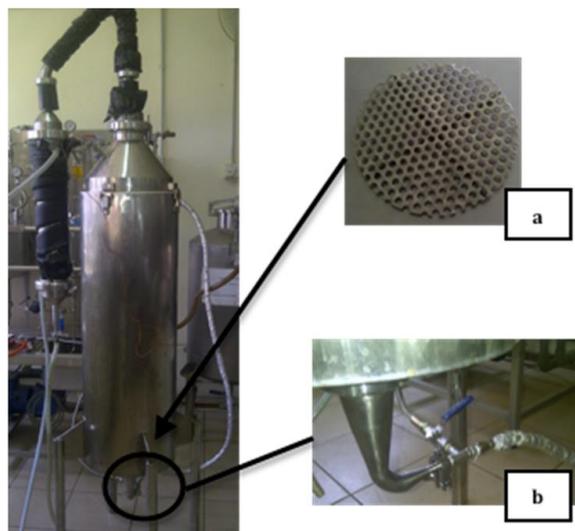


Fig. 4. The steam distillation unit: (a) perforated grid plate, (b) connection to a boiler or steam inlet

Chemical Properties

Gas chromatography (GC) analysis was performed using a Hewlett-Packard 6890 device equipped with a split/splitless injector (250 °C, split ratio 100:1) using an Agilent HP-5MS column (30 m × 0.25 mm, df: 0.25 μm). The temperature program started at 70 °C (5 min) and rose to 325 °C at a rate of 3 °C/min. The injector and detector temperatures were both 250 °C. Helium was used as the carrier gas at a flow rate of 1 mL/min. The injected sample consisted of 1.0 mL of pure citronella oil.

Gas chromatograph-mass spectrometry (GC-MS) was performed using a Hewlett-Packard 5973/6890 system operating in EI mode (70 eV), equipped with a split/splitless injector (250 °C, split ratio 100:1) using an Agilent HP-5MS column (30 m × 0.25 mm, df: 0.25 µm). The temperature was programmed to start at 70 °C (5 min) and increased to 325 °C at a rate of 3 °C/min. The injector and detector temperatures were both 250 °C. Helium was used as the carrier gas at a flow rate of 1 mL/min.

The identification of the chemical constituents was carried out by calculating the retention indices and by comparing with the peaks of the Wiley and Adams library search data. A comparison of the GC-MS analyses of citronella oil samples obtained using the different methods was conducted with citronella oil produced by steam distillation at MARDI Kuala Linggi as a reference oil.

Physical Properties

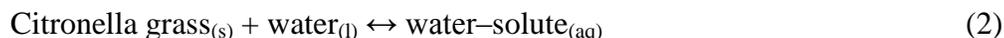
The specific gravity, refractive index, and colour were determined for the citronella oil samples from the different extraction methods and the reference oil. The specific gravity of the oil was determined by a Densitometer DA-130N. The refractive index of the oil was determined using an Atago No. 68534, and the colour of the oil was analysed using a Lovibond Tintometer Model E.

Scanning Electron Microscopy (SEM) Properties

SEM micrographs of the citronella grass raw material and citronella samples from different extraction methods were obtained with a Hitachi scanning electron microscope Model S-3400N under high-vacuum conditions and at an accelerating voltage of 15 kV. The SEM images were digitally recorded as high-resolution topographic images at 500× magnification.

Proposed Kinetic Model

To date, few reports are available in the literature regarding the kinetics of ohmic-heated hydro-distillation (Gahavian *et al.* 2011, 2012), and there is no report concerning which model could be used to verify the reaction kinetics of ohmic-heated hydro-distillation. Hence, a second-order mechanism was assumed to be a plausible model for the kinetics of ohmic-heated hydro-distillation, as well as hydro-distillation and steam distillation. A second-order kinetics model means that the extraction occurred in two simultaneous processes. The amount of extracted oil increased rapidly at the beginning of extraction and decreased gently until the end of the extraction process (Ho *et al.* 2005; Rabesiaka *et al.* 2007; Meizane and Kadi 2008; Uhm and Yoon 2011). The extraction was assumed to proceed as (Ho *et al.* 2005),



assuming that the operation was only a function of the water-solute in suspension because the activities of the solid (citronella grass) and the pure liquid (water) were unity. The concentration level of saturated water-solute in suspension is considered constant for the same extraction conditions. After the water-solute had dissolved into suspension, the derivatives diffused from the porous citronella grass particles into the bulk liquid phase. The extraction order and rate constant must be determined experimentally. Therefore, a second-order rate law expression for the extraction, which demonstrated how the rate depends on the water-solute dissolved to suspension, was employed.

The second-order kinetic model was as follows (Ho *et al.* 2005):

$$dC_t/dt = k (C_s - C_t)^2 \quad (3)$$

where k is the second-order extraction rate constant (kg.kW/mL), C_s is the concentration of oil at saturation (mL/kg.kWh), and C_t is the concentration of oil in the solution (mL/kg.kWh) at any time t (min). The rate of dissolution for the oil contained in the solid can be described by this equation.

The integrated rate for a second-order extraction can be obtained by considering the boundary conditions $t = 0$ and t from $C_t = 0$ to C_t :

$$C_t = \frac{C_s^2 kt}{1 + C_s kt} \quad (4)$$

The linear form of Eq. 4 is:

$$\frac{t}{C_t} = \frac{1}{kC_s^2} + \frac{t}{C_s} \quad (5)$$

The extraction rate can be written as follows:

$$\frac{C_t}{t} = \frac{1}{[(1/kC_s^2) + (t/C_s)]} \quad (6)$$

The initial extraction rate, h (when t approaches 0), can be written as follows:

$$h = kC_s^2 \quad (7)$$

Rearranging Eq. 6, the concentration of the oil at any time can be obtained from:

$$C_t = \frac{t}{[(1/h) + (t/C_s)]} \quad (8)$$

By plotting $t/[C_t/P]$ versus t , the initial extraction rate (h), the extraction capacity (C_s), and the second-order extraction constant (k) can be calculated experimentally.

RESULTS AND DISCUSSION

Model Analysis

The essential oil extracted (with respect to the total power input) by ohmic-heated hydro-distillation was substantially higher (at 1.911 mL/kg.kWh) when compared to those obtained by steam distillation, at 0.880 mL/kg.kWh. As shown in Fig. 5, all extracted oils were fully recovered after 2 h, 1 h, and 1.5 h for ohmic-heated hydro-distillation, hydro-distillation, and steam distillation, respectively.

The experimental results for the three different methods were plotted using a second-order model, by plotting $t/[C_t/P]$ versus time, as shown in Fig. 6. Overall, the oil yield increased rapidly at the beginning of the extraction and became slower thereafter.

Changes in solute concentration in the liquid phase affected the mass transfer of the extraction process (Pin *et al.* 2011).

In the first stage, the oil concentration was low. Thus, the oil diffused rapidly from the solute to the liquid phase because of the mass transfer effect. Further, the free oil on the surface of the citronella grass was solubilised when the solute was exposed to fresh solvent (Sayyar *et al.* 2009).

However, in the second stage, the diffusion rate decreased along with the increase in extraction time due to the high solute concentration in the liquid. Prolonged extraction time (after achieving the maximum level) did not yield any significant increase in the amount of oil extracted.

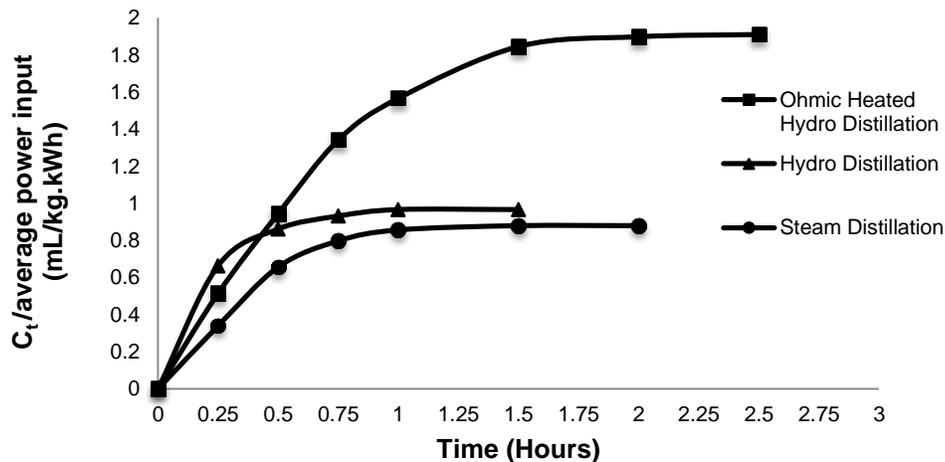


Fig. 5. Extracted oil yield with respect to the total power input

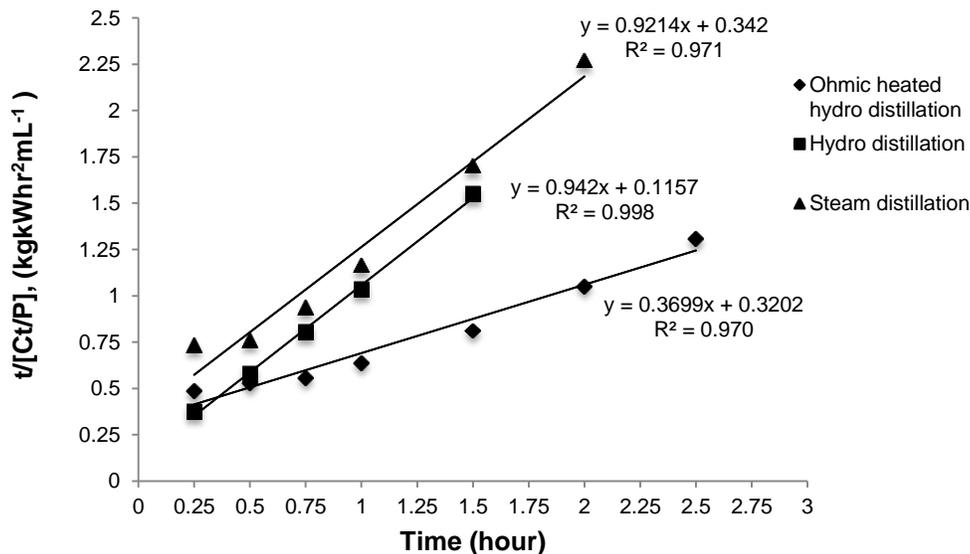


Fig. 6. Second-order extraction kinetics of citronella oil for the different extraction methods

The straight line exhibited in Fig. 6 indicated good agreement with the second-order extraction model with the experimental results. This agreement indicated that the three different methods of citronella oil extraction all behave in two successive stages. The initial extraction rate (h), extraction capacity (C_s), second-order extraction constant

(k), and coefficient of determination (R^2) were calculated experimentally by referring to the linear plot in Fig. 6. From the graph of $t/[C_i/P]$ versus time, the slope was equal to $1/C_s$ and the intercept was equal to $1/kC_s^2$. The data are shown in Table 1.

Table 1. Linearisation of Second-order Kinetic Model for Different Extraction Methods

Extraction method	Time to reach boiling point (min)	Extraction time (min)	Power input (kWh)	Yield (mL/kWh)	Linearisation of second-order kinetic model			
					Concentration of oil at saturation, C_s (mL/kg.kWh)	Second-order extraction rate, k (kg.kW/mL)	Initial extraction rate, h (mL/kg.kW.h ²)	R^2
Ohmic-heated hydro-distillation	38 min	120 min	2.329	7.640	2.703	0.427	3.120	0.970
Hydro-distillation	32 min	60 min	4.290	3.870	1.062	7.633	8.643	0.998
Steam distillation	35 min	90 min	4.260	1.690	1.085	2.484	2.924	0.971

As shown in Table 1, the time it takes to reach the boiling point by different extraction methods was obtained within the range of 32 to 38 min. Theoretically, the rate constant (k) and the initial extraction rate are essential in order to quantify the speed of the chemical reaction (Wright 2004). The higher rate constant indicating the reaction proceeds faster. It was observed that the extraction rate constant (k) and the initial extraction rate (h) for hydro-distillation were the highest; but in contrast, the concentration of oil at saturation was the lowest. During hydro-distillation, the oil-water solution permeates through swollen membranes only *via* osmosis, and as the temperature between the solid and liquid material equilibrates, the aqueous phase of citronella-water mixture will begin to boil at the boiling temperature of water. Finally, the generated steam carries away the vaporised oil, which is then condensed in the condenser (Handa 2008). The lowest extracted oil yield for hydro-distillation could presumably be due to the requirement for large quantities of steam; this would lead to less oil that could completely vaporise until the end of extraction.

In terms of extracted oil yield *versus* power input, it can be stated that ohmic-heated hydro-distillation resulted in 7.64 mL/kWh more oil yield in comparison to hydro-distillation and steam distillation. This is supported by the ohmic-assisted hydro-distillation of essential oils from *Zataria multiflora* Boiss., which has been shown to yield 6.67 mL/kWh, more than hydro-distillation, which produced only 1.82 mL/kWh oil (Gahavian *et al.* 2011). Additionally, in another comparative study of ohmic-assisted hydro-distillation with traditional hydro-distillation for the extraction of essential oils from *Thymus vulgaris* L. (Gahavian *et al.* 2012), for 1 kWh of power consumption, the

extracted oil yield was 0.76 mL for hydro-distillation and 2.56 mL for ohmic-assisted hydro-distillation.

Kinetics data for the three different extraction methods were modelled and verified using simple linear regression. The proposed second-order kinetics model fitted well with the experimental data. Based on the overall coefficients of determination, R^2 more than 0.97, the proposed model successfully predicted the relationship among the independent variables on citronella oil extraction. In fields where random fluctuations are of smaller magnitudes (for instance, engineering), coefficients of determination of less than 0.95 may imply an unsatisfactory fit with the experimental data (Freund *et al.* 2010). In summary, the model used was judged to be a satisfactory fit with the experimental data.

Chemical Analysis

The GC-MS analysis of citronella oil led to the identification of different constituents, representing four sources of citronella oil. The identified constituents are listed in Table 2. In total, 13 constituents were identified. Among them, 13 were identified in the essential oil extracted by ohmic-heated hydro-distillation, 9 were from hydro-distillation, and 11 were from both steam distillation and the reference oil. It was found that 8 constituents were the same: citronellal, citronellol, neral, geranial, geraniol, citronellyl acetate, eugenol, and geranyl acetate. The most abundant constituents were citronellal, citronellol, and geraniol. Prior studies have also reported that these are the main constituents of citronella oil (Laksmono *et al.* 2007; Akhila 2010; Nor Azah *et al.* 2002; Weiss 1997).

Table 2. Chemical Constituents of Citronella Oil

Chemical constituents	Ohmic-heated hydro-distillation (%)	Hydro-distillation (%)	Steam distillation (%)	Reference oil (%)
linalool	1.63	-	-	1.44
citronellal	19.61	22.09	20.99	13.11
1-ethyl -2-methylcyclopentene	0.43	-	-	-
4-nonanone	0.76	0.80	0.63	-
citronellol	12.97	12.26	13.64	11.97
neral	6.60	7.97	4.94	6.84
geraniol	41.15	41.48	42.43	42.38
geranial	6.42	8.39	4.39	9.55
citronellyl acetate	1.75	1.37	2.13	1.50
eugenol	1.06	0.91	0.78	1.85
geranyl acetate	5.66	4.73	7.74	4.93
trans- β -caryophyllene	0.69	-	0.97	1.97
γ -cadinene	1.26	-	1.35	4.46

Chemical analysis revealed the main constituents of citronella oil (%) from the different methods, and results are shown in Fig. 7. The percentages of citronellol and geraniol were almost the same in the four sources of citronella oil. Conversely, citronellal compound was in small proportion in the citronella reference oil as compared to the oil extracted from the other extraction methods used. The chemical profile or essential oil composition may be affected by environmental conditions, geographic variations, genetic factors and evolution, political/social conditions, time, and technique of harvesting

(Singh *et al.* 1996; Dhar *et al.* 1996; Figueiredo *et al.* 2008; Costa *et al.* 2005). In this study, the raw samples used were obtained from the same sources. Therefore, the quantitative differences in the citronella oil composition could be due to the extraction processes used.

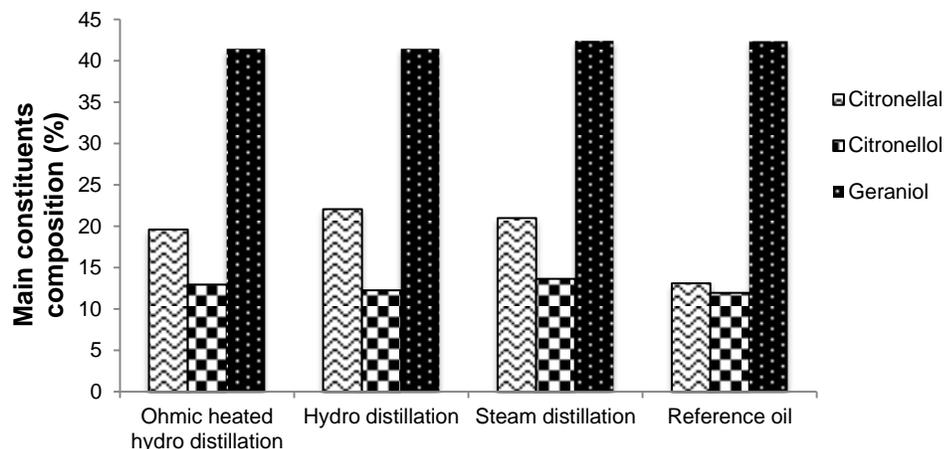


Fig. 7. Main constituents of citronella oil (%) from different extraction methods

Physical Analysis

The physical properties (refraction index at 20 °C, specific gravity at 20 °C, and colour) of citronella oil extracted by ohmic-heated hydro-distillation, hydro-distillation, steam distillation, as well as the reference oil, are shown in Table 3.

Table 3. Physical Properties of Citronella Oil from Different Sources

Extraction method	Refraction index at 20 °C	Specific gravity at 20 °C	Colour
Ohmic-heated hydro-distillation	1.4671	0.8879	0.7R 4Y
Hydro-distillation	1.4671	0.8888	2.0Y
Steam distillation	1.4671	0.8896	0.5R 3Y
Reference oil	1.4743	0.8916	0.7R 4Y

The refraction index and specific gravity are two physical parameters that provide beneficial information about the purity of citronella oil. Based on the physical analysis, the refraction index and specific gravity at 20 °C for all four different sources of citronella oil were about 1.47 and 0.89, respectively. Colour is an important feature that determines the consumer's acceptability of citronella oil products. Normally, commercial citronella oil is yellow in colour. In this work, the intensity of yellow and red was measured. The citronella oil extracted by ohmic-heated hydro-distillation and the reference citronella oil from MARDI were 0.7R 4Y. The extracted citronella oil by hydro-distillation exhibited colour in the range of 2.0Y, whereas the extracted citronella oil by steam distillation exhibited colour in the range of 0.5R 3Y.

The characteristics of the colour red were determined in the citronella oil extracted using ohmic-heated hydro-distillation, steam distillation, and the reference oil.

The red colour was due to trans- β -caryophyllene and γ -cadinene constituents in the citronella oil (Abena *et al.* 2007; Harjeet *et al.* 2011). Vlora (2009) also determined that the essential oil obtained from the red leaves of the American sycamore (*Platanus occidentalis*) showed a high concentration of bicyclogermacrene, β -caryophyllene, and γ -cadinene. However, there was no red colour found when using the hydro-distillation method. The appearance of red colour when using other extraction methods also may be caused by lipid oxidation occurring in the extraction systems. Erwin *et al.* (2004) mentioned that the oxidation mechanism is based on lipoxygenase activity. The lipoxygenase enzyme will form hydroperoxides or iron atom during oil extraction. Metal ions such as Fe and Cu play a major role in the initiation of this autoxidation process.

Scanning Electron Microscopy (SEM) Analysis

Figure 8 shows images of citronella grass obtained by scanning electron microscopy, showing the appearance before and after the extraction processes. Figure 8(a) is a micrograph of the untreated citronella (raw material before extraction). Figures 8(b-d) show scanning electron microscope images of citronella that had undergone ohmic-heated hydro-distillation, hydro-distillation, and steam distillation, respectively.

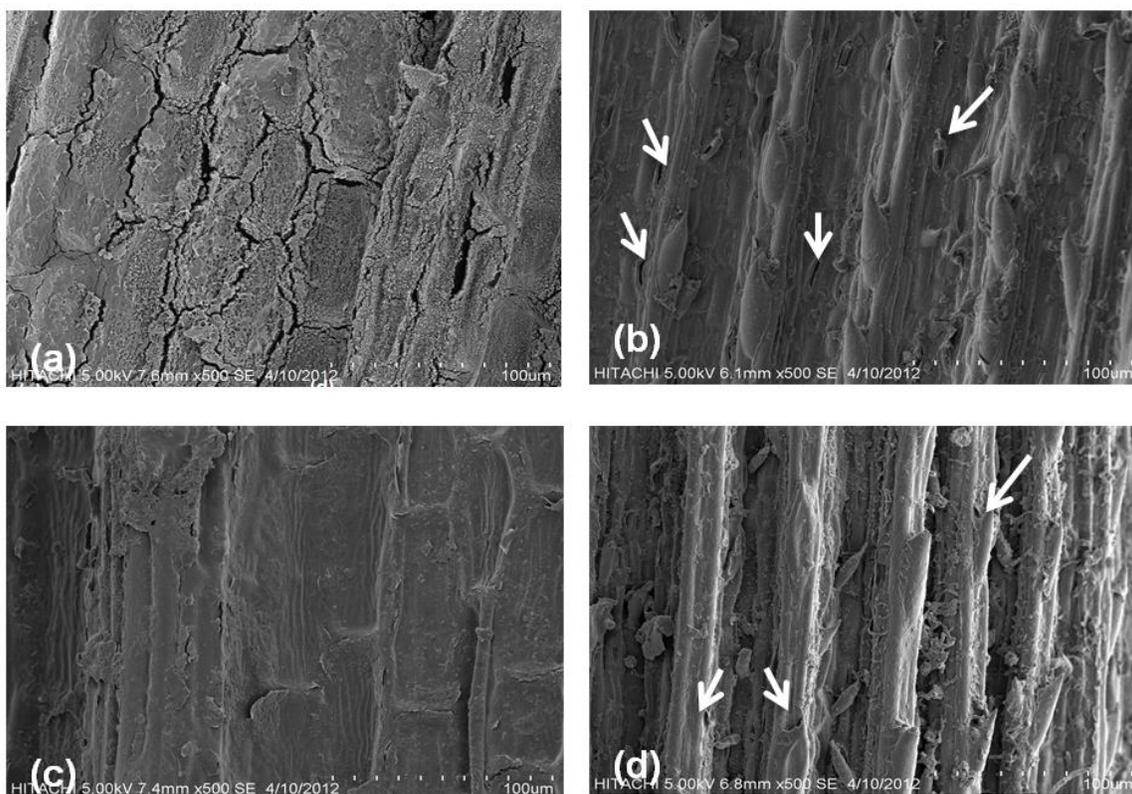


Fig. 8. SEM of citronella grass: (a) raw material, (b) ohmic-heated hydro-distillation, (c) hydro-distillation, (d) steam distillation

The citronella that had undergone ohmic-heated hydro-distillation and steam distillation showed some microfractures, but less cell wall degradation than hydro-distillation. In addition, after ohmic-heated hydro-distillation, the cell walls were less rigid compared to citronella extracted with steam distillation. The cells of the hydro-distillation sample were less dense and exhibited pronounced swelling, but did not show

any microfractures. Additionally, the hydro-distillation method resulted in clear cell wall degradation and less lignin in comparison with the other two extraction methods. Secretory cavities (arrows in Fig. 8(b) and 8(d)) were scattered throughout the surface of the citronella from ohmic-heated hydro-distillation and steam distillation. Secretory (or schizogenous) cavities are the specific plant tissues of citronella that produce and store citronella oil (Handa 2008).

During hydro-distillation, the cells tended to swell only within the extraction time, but they did not burst. This situation might be insufficient for the release of citronella oil within the cell lignin. Turner *et al.* (1998) and Boz *et al.* (2009) mentioned that the schizogenous secretory cavities invariably swell and often burst because of osmotic swelling of the glandular cells. This is attributed to the heat transfer that occurs only through conduction and convection (Golmakani and Rezaei 2008). Hence, pre-treatments or assisted methods have been proposed to facilitate the rupture of the cell walls, thus producing further bursts and creating schizogenous cavities or pores with the hydro-distillation process.

In contrast, ohmic-heated hydro-distillation opened the pores of the schizogenous cavities to release the oil. This phenomenon can be related to the formation of transient pores in the cell membranes by a process called electroporation. This enabled the transport of molecules and ions, known as the electro-osmosis effect, which increased the diffusion of materials (Lakkakula *et al.* 2004; Pavlin *et al.* 2007; Knirsch *et al.* 2010). Additionally, in ohmic-heated hydro-distillation, the heat is generated volumetrically within the material. Thus, heat penetration into the materials and other areas could occur rapidly. This agreed with Fourier's Law, whereby a high local temperature gradient and a small particle dimension result in a high local heat flux (Parrott 1992; Gahavian *et al.* 2012).

A similar finding was obtained for the citronella sample using the steam distillation method. The appearance of pores in the SEM image indicated that direct steam distillation tended to burst the sacs containing the oil molecules, leading to oil production from the cells (Chemat 2010). However, direct steam distillation was operated at atmospheric pressure, which might not be enough to extract the citronella oil. This phenomenon may be one of the reasons that the cell walls of citronella extracted by steam distillation were more rigid than those from ohmic-heated hydro-distillation. Occasionally, it was necessary to use steam at a higher pressure to obtain a good yield. Silva *et al.* (2011) mentioned that better extraction efficiency of citronella oil by supercritical CO₂ was obtained under high pressure. The best conditions were found to be 353.15 K and 18.0 MPa. A higher pressure caused the cell wall to rupture or produce an enlargement of pores so that more steam was allowed to pass through the pores (Vinatoru *et al.* 1999), leading to the diffusion of much more oil from the cells.

Overall, the structural changes in the citronella grass cell walls elucidate the mechanism of the various extraction methods. The different heating mechanisms of the different citronella oil extraction methods resulted in varied recovery of secondary metabolites and other aroma constituents that naturally occurred in the cell walls or cytoplasm. As the cell wall disrupted, it facilitated the mass transfer of the solvent into the plant material and the secondary metabolites into the solvent. The diffusion path could be shortened, thus increasing the extraction efficiency (Destandau *et al.* 2013).

CONCLUSIONS

1. Ohmic-heated hydro-distillation is superior to hydro-distillation and steam distillation in terms of oil yield. Ohmic-heated hydro-distillation (7.64 mL/kWh) resulted in about two times the oil yield of hydro-distillation (3.87 mL/kWh) and four times the oil yield of steam distillation (1.69 mL/kWh). The proposed second-order kinetic model fitted well with the experimental data, with a coefficient of determination, R^2 , that was more than 0.97.
2. The citronella oils obtained by the three methods were similar in their main constituents, namely, citronellal, citronellol, and geraniol. The results for specific gravity and refractive index at 20 °C ranged from 0.8879 to 0.8916 and from 1.4671 to 1.4743, respectively. The citronella oil colour intensity in terms of red and yellow units ranged from 2.0Y to 0.7R and 4Y.
3. The SEM images of citronella grass reveal that the lignocellulosic sources of the extracted citronella oil were schizogenous cavities within the cell lignin of citronella grass.
4. SEM images of the citronella grass from the ohmic-heated hydro-distillation method indicated that ohmic heating led to the formation of transient pores in the cell membranes, even though the cell walls were still rigid. Ohmic-heated hydro-distillation is feasible as an alternative method of citronella oil extraction.

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