

Full Length Research Paper

A practical study for new design of essential oils extraction apparatus using ohmic heating

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Ohmic heated hydro-distillation (OHHD) apparatus was designed and manufactured in the food engineering Laboratory, Agriculture College, Basrah University. It consists of poles, glass condenser, plastic cylinder and voltage regulator. OHHD at 60, 70 and 80 V and hydro-distillation (HD) (conventional method) are compared and evaluated in extraction time, input power, energy consumption of essential oil, heating rate, essential oil yield, predicted yield, specific gravity, reflective index and chemical composition by using GC-MS. Eucalyptus oil was extracted from dried Eucalyptus leaves using OHHD at 60, 70, 80 V and HD. The results showed that OHHD at 60, 70, 80 V and HD have the extraction time of 110, 90, 70 and 142 min. respectively, and essential oil yield were 1.64, 1.90, 2.19 and 1.36 % respectively. Also, the energy consumption of essential oil by using OHHD at 60, 70 and 80 V was significantly ($p < 0.05$) lesser than HD. As well as, the input power (kW) at using OHHD was lesser than HD. The higher heating rate was by using OHHD at 80 V. Fick's second law of one-dimensional unsteady-state diffusion was applied, and both transfer kinetic coefficients increased with increasing applied voltage. However, there is no significant differences among OHHD at 60, 70, 80 V and HD for the specific gravity and reflective index. GC-MS analysis of the Eucalyptus leaves essential oil revealed the presence of 29, 30, 27 and 28 compounds from the OHHD at 60, 70, 80 V and HD respectively.

Key words: Ohmic heating, eucalyptus leaves, essential oil, hydro-distillation, oil yield.

INTRODUCTION

Eucalyptus oil is readily steam distilled from the leaves and can be used for cleaning and as an industrial solvent, as an antiseptic, for deodorizing and in very small quantities in food supplements, especially cough drops, sweet, decongestants and toothpaste. It has bug repellent properties (Jahn 1991), and is an active ingredient in some commercial mosquito repellents (Fradinand and Day, 2002). Eucalyptus globules are the principal source of eucalyptus oil worldwide.

Essential oil or etheric Oil means a volatile oil and is obtained from plants by steam distillation method (Guenther, 1987). Öztekin and Martinov (2007), stated that essential oils are used in medicinal and pharmaceutical purposes, food and food ingredients, herbal tea, cosmetics, perfumery, aromatherapy, pest and disease control, gelling agents, dying in fabrics, plant growth regulators and paper making.

Essential oils often extracted from the flowers, leave and roots plants are a good solution to this problem. The distillation methods have advantages of extracting pure and refine essential oils by evaporating the volatile

essence of the plant material (Malle and Schmickl, 2005). Essential oils are extracted from various parts of the plant such as bark, leaves, roots, wood, seeds or fruits, flowers, burgeons, branches or all plants (Öztekin and Martinov, 2007). About 65% of the essential oils produced in the world are obtained from the woody plants that are trees and bushes (Baser, 1999). Herbage products have been marketed as fresh or dry products or as essential oils. These are in general used as raw materials for the extraction of active substances or chemical precursors and mainly for the production of teas, homemade-remedies, fluid extracts and also powders resulting from dried and comminuted plants or from the drying an extract (Runha et al., 2001).

There are several techniques for isolating volatiles from plant materials are: water distillation, steam distillation, water and steam distillation, hydro-diffusion, solar distillation, microwave distillation, supercritical extraction, pressing, extraction with solvent extraction, extraction with fat. Water distillation is generally used to extract the essential oils of the dried or powdered plant materials,

during the boiling process, the volatile component essential oils from special structures inside or diffusion mechanism material, is mostly extracted at temperature just below 100°C by at the surface of the plant (Öztekin and Martinov, 2007). Asolar distillation system was developed using Scheffler fixed-focus concentrator but it worked effectively only during sunny days (Munir and Hensel, 2009).

Water and steam distillation is a method that has characteristics of both water distillation and steam distillation. With this method, a metal grid is placed in the still above the level of water and the plant material is thus avoided (Oyen and Dung, 1999). The design of the equipment used is generally very similar to that used in water distillation (Lawrence, 1995).

Indirect steam distillation a separate steam generator is attached to the still. As in steam and water distillation, plant material is placed on a grid in the distillation vat, but no water is added. Steam produced in the generator is forced through the material to be distilled. High pressures steam is often used e.g. Steam of 5-10 bars pressure at 150-200°C. (Lawrence, 1995). However, these techniques have several disadvantages, such as low yield, time consuming process, loss of volatile compounds, degradation of thermo-sensitive compounds and few adjustable parameters to control the process selectivity. Therefore, the search for extraction techniques with high efficiency and selectivity is desirable (Ghsemi et al. 2011). Furthermore, elevated temperatures can cause partial or full degradation of natural constituents especially monoterpenes which are vulnerable to structural changes under steam distillation conditions. Conventional solvent extraction method is likely to involve losses of more volatile compounds during removal of the solvent (Moyler, 1991; Presti et al., 2005).

Supercritical fluid extraction (SFE) is an alternative sample preparation method with general goals of reduced use of organic solvents and increased sample throughput.

The factors to consider include temperature, pressure, sample volume, analyte collection, modifier (co-solvent) addition. A weakness of supercritical extraction process is the price of equipment which is very expensive due to high requirement of the operating pressure (Ramadhan et al., 2010).

Finally, expression or cold pressing, as it is also known, is used in the production of citrus essential oils. The term expression indicates any physical process in which the essential oil glands in the peels are crushed or chopped to liberate the oil.

On the other hand, Abdull-Neby (2009), stated that the highest percentage of essential oils was found in cardamom which reached to 4 %, while the essential oils as eucalyptus, lemon and orange reached to 3.5%, 0.82% and 0.75% respectively, by using water distillation method.

Ohmic heating is a thermal process in which heat is internally generated by the passage of Alternating electrical

Current (AC) through a body such as a food that convert as an electrical resistance (Shirsat et al., 2004). The main advantages of ohmic processing are the rapid and relatively uniform heating achieved (Zareifard et al., 2003). In addition AC voltage is applied to the electrodes at both ends of the product body.

The rate of heating is directly proportional to the square of the electrical field strength and the electrical conductivity. The electric field strength can be varied by adjusting the electrode gap or the used voltage. Ohmic-hydro-distillation is an advanced hydro-distillation technique using ohmic heating process and could be considered as a novel method for the extraction of essential oils. As well as, the results of this study introduced as a verdant technology because of less power required per ml of essential oil extraction. (Gavahian et al., 2012).

The aim of this study is design of new apparatus for extraction of essential oils from Eucalyptus leaves by using Ohmic heating.

MATERIALS AND METHODS

Ohmic extractor

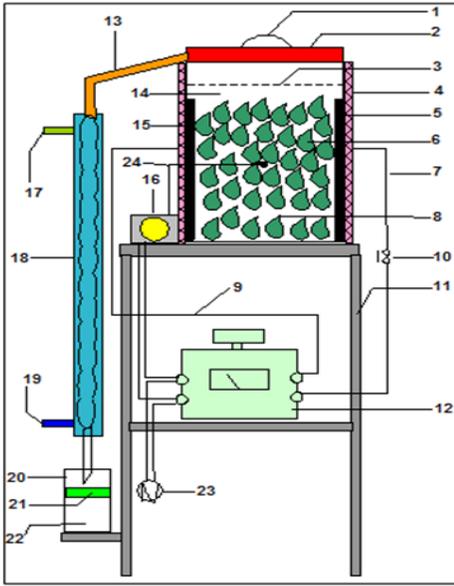
The ohmic heated hydro-distillation (OHHD) apparatus was designed and manufactured in the food engineering Laboratory, Agriculture college, Basrah university, Iraq. It consist of two electrodes (poles) (5) of 0.075×0.14 m which made from stainless steel 316, inner plastic cylinder (14) of 0.14 m diameter and 0.195 m height, it's capacity 150 g of dried plants (6). Inner plastic cylinder (14) was put in the outer plastic cylinder and the gap between them is 0.025 m, plastic cover (2), condenser (18) made of glass, condensate water and oil container (20) and voltage regulator (12), Figures 1 and 2.

Hydro-distillation

150 g of dried Eucalyptus leaves were hydro-distilled with 600 ml of distilled water using hydro-distillation method that is used as a reference for the quantification of essential oils.

Eucalyptus Leaves

Eucalyptus leaves were obtained from Agriculture researches station in the Agriculture College, Basrah University. Eucalyptus leaves were then dried under ambient conditions (30-35 °C) for three days in a large screened tray. The moisture content of the Eucalyptus leaves was measured in triplicate using a laboratory oven by drying until constant weight according to AOAC (1980). The Eucalyptus leaves were cut approximately 0.25 cm by a knife then put in the ohmic extractor.



1. Handle, 2.cover, 3.water level, 4.insulator, 5.and 15.electric poles, 6.plants, 7.wire, 8.water, 9.wire, 10.electric switch, 11.wood body, 12.voltage regulator, 13.plastic pipe, 14.cylinder, 16.temperature indicator, 17.hot water outlet, 18.heat exchanger (condenser), 19.cold water inlet, 20.flask, 21.essential oil layer, 22. Collected water, 23.AC voltage supply, 24.Thermocouple.

Figure 1. Schematic diagram for the essential oils extraction apparatus using Ohmic heating.



Figure 2. A photograph of the locally manufactured ohmic extractor.

Physical properties

The refractive index and specific gravity were determined

for the Eucalyptus leaves oil samples for different extraction methods (ohmic-heated hydro-distillation extractor at 60, 70 and 80V and hydro-distillation extraction). were measured according to food chemical codex (FCC, 1996) at 25 °C.

The system Performance Coefficient (SPC) is calculated from Equation (1) (Icier and Ilicali, 2004; 2005):

$$SPC = \frac{Q_t}{E_g} \quad (1)$$

$$E_g = Q_t + E_{loss} = \sum \Delta VIt \quad (2)$$

$$Q_t = mC_p (T_f - T_i) \quad (3)$$

Where, M: mass (kg), T_f : final temperature (°C), T_i : primary temperature (°C), E_g : amount of given energy (J) and Q_t : the amount of taken heat (J).

The specific heat was obtained as follows: (Toledo, 2007)

$$C_{pw} = 4176.2 - 9.0864 \times 10^{-3}T + 5473.1 \times 10^{-6}T^2 \quad (4)$$

The heat loss to the surroundings by natural convection has been calculated from the following equation: (Icier and Ilicali, 2005).

$$E_{loss} = \bar{h}\pi DL(\bar{T}_w - T_{amb.})\Delta t \quad (5)$$

The average of heat transfer coefficient was calculated from the following equation (Geankoplis, 1993):

$$\bar{h} = 1.32 \left[\frac{\Delta T}{D} \right]^{1/4} \quad (6)$$

Where C_{pw} is the specific heat (J/kg.K), h is the heat transfer coefficient (W/m^2K), D , is the outer diameter of the cylinder (m), T_w is the outer wall temperature (°C), $T_{amb.}$ is the ambient temperature (°C), E_{loss} is the heat loss by natural convection (J) and ΔT is the average temperature driving force calculated from the initial and final outer wall temperatures and the ambient temperature.

In which the electric field strength, E (V/cm), is calculated by Equation (7) (Floury *et al.*, 2006):

$$E = \frac{V}{L} \quad (7)$$

Electrical conductivity is calculated according to Equation (8) (Wang and Sastry, 1993; Iciier *et al.*, 2008):

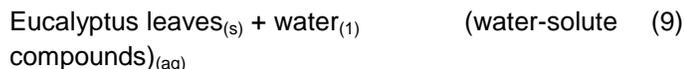
$$\sigma = \frac{IL}{VA} \quad (8)$$

I : current (A), L : distance between poles (m), V : voltage (V), A : section area (m^2) and σ : electrical conductivity (S/m).

Mathematical modeling

Gahavian *et al.* (2012), stated that few reports are available in the literature concerning the kinetic ohmic-heated hydrodistillation. A second order mechanism was

assumed to be an accepted model for the kinetic of ohmic- heated hydro-distillation and hydro-distillation. Ho et al.(2005);Rabesiaka et al. (2007); Meizane and Kadi (2008);Uhm and Yoon (2011), announced that the amount of extracted oil increased rapidly at the beginning of extraction and significantly ($p<0.05$) decreased until the end of the extraction process. The extraction was simply assumed to process as:



Assuming that the operation is only a function of water-soluble compounds in suspension because the activities of the solid (Eucalyptus leaves) and the pure liquid (water) are unity. The mass transfer of essential oil through plant particles during ohmic-hydrodistillation occurs as unsteady state distillation. Fick's second law of one-dimensional unsteady-state diffusion can be applied as a following equation:

$$\frac{\partial q_p}{\partial t} = D_{eff} \frac{\partial^2 q_p}{\partial x^2} \quad (10)$$

Where q_p is the average concentration of essential oil in the plant particles at time t , x is the distance along the direction of diffusion and D_{eff} is the effective diffusion coefficient. Integration equation (10) according to (Treybal,1975) leads to equation (11):

$$\frac{(q_o - q)}{q_o} = A e^{-kt} \quad (11)$$

Where q is the average yield of Eucalyptus leaves oil at time t , q_o is the initial average concentration of Eucalyptus leaves oil, A is a constant and k is a kinetic constant including the effective diffusion coefficient. A and k were calculated by using the solver in the excel program depended on a various statistical parameters such as the reduced chi-square (χ^2) as the mean square of the deviation between the experimental and predicted values for the model and Root Mean Square Error analysis (RMSE) and coefficient of determination (R^2). The parameter can be calculated as following Equations (12 to 15):

$$R^2 = \frac{\sum_{i=1}^n (q_{pre.} - \overline{q_{pre.}})^2}{\sum_{i=1}^n (q_{exp.} - \overline{q_{exp.}})^2} \quad (12)$$

$$\chi^2 = \frac{\sum_{i=1}^n (q_{exp.} - q_{pre.})^2}{N - n} \quad (13)$$

$$RMSE = \sqrt{\frac{\sum_{i=1}^N (q_{pre.} - q_{exp.})^2}{N}} \quad (14)$$

$$RE\% = \left| \frac{\sum_{i=1}^N (q_{pre.} - q_{exp.})}{N} \right| \times 100 \quad (15)$$

Where $q_{exp.}$ is the experimentally observed moisture ratio. $q_{pre.}$ Is the predicted moisture ratio, N is the total number of observation, n is the number of constants in the model and RE % is the error percentage. Extracted oil yield: the extracted oil was determined at 2 min. intervals until it reached equilibrium. The end of each interval, the extracted oil yield was estimated as follows:

$$\text{Yield (\%)} = \frac{W_{eo}}{W_{is}} \times 100 \quad (16)$$

Where W_{eo} is the weight of Eucalyptus leaves oil collected (g) and W_{is} is initial weight of sample (g).

Heating rate (H_r) can be estimated from the equation (17) (Halleux et al., 2005):

$$H_r = \frac{T}{t} \quad (17)$$

GC-MS

GC-MS analysis was carried out using GCM-QP2010 Ultra, Shimadzu type. The operation conditions are illustrated in table 1.

STATISTICAL ANALYSIS

All extraction treatments (Ohmic heated extraction at 60, 70, 80 V and Clevenger apparatus (hydro-distillation HD)) were performed in triplicates. ANOVA was performed to determine significant differences among means and LSD test was used to compare among the means using SPSS program (SPSS, 2009).

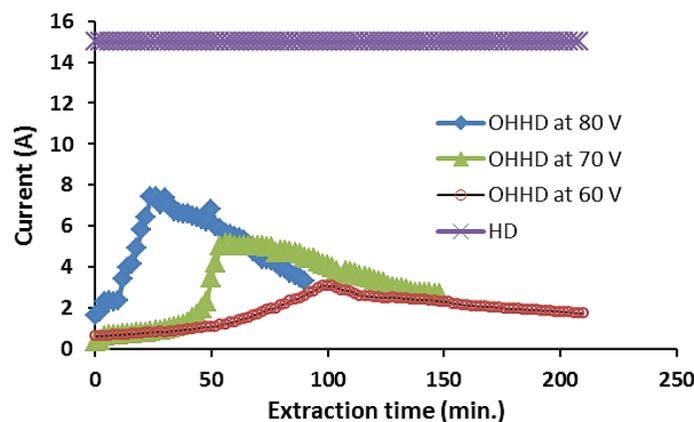
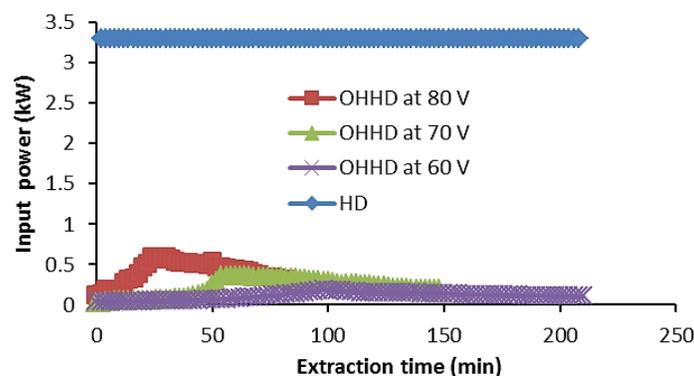
RESULTS AND DISCUSSION

Figure 3. Illustrate changes in current during time at different applied voltages by ohmic heating for Eucalyptus leaves. The current was increased significantly ($p<0.05$) with increasing extraction time and reached to the maximum value (7.45 A) after 24 minutes when using the OHHD at 80 V, and then decreased thereafter. The reason for this decline is the reducing level of water between the poles as a result of vaporization. On the other hand, the lower value of the current was using the OHHD at 70 V then OHHD at 60 V 5.13, 2.98 A respectively. While HD needs 15A.

As shown in Figure 4 the power input was increased with increasing extraction time up to maximum value at boiling point then dropped after that for OHHD at 60, 70 and 80 V. This because of mixture evaporation during boiling led to reducing mixture level between poles. Also,

Table 1. The operation conditions of GC-MS.

[GC-2010]	[GC Program]
Column Oven Temp. :40.0 °C	[GCMS-QP2010 Ultra]
Injection Temp. :280.00 °C	Ion Source Temp :200.00 °C
Injection Mode :Split	Interface Temp :280.00 °C
Flow Control Mode :Pressure	Solvent Cut Time :3.00 min
Pressure :96.1 kPa	Detector Gain Mode :Relative
Total Flow :56.0 mL/min	Detector Gain :0.70 kV +0.10 kV
Column Flow :1.71 mL/min	Threshold :0
Linear Velocity :47.2 cm/sec	SIF\$(-Group 1 - Event 1-)
Purge Flow :3.0 mL/min	Start Time :3.00min
Split Ratio :30.0	End Time :28.00min
High Pressure Injection :OFF	ACQ Mode :Scan Event
Carrier Gas Saver :OFF	Time :0.50sec
Splitter Hold :OFF	Scan Speed :1666
	Start m/z :50.00
	End m/z :800.00
	⇒

**Figure 3.** Changes in current during time at different applied voltages by ohmic heating for Eucalyptus leaves**Figure 4.** Power input vs. extraction time at different applied voltages by ohmic heating for Eucalyptus leaves.

the maximum value of input power was 0.512 kW when using OHHD at 80 V. On the other hand, HD needs 3.3 kW. However, HD needs power input more than OHHD at 60, 70 and 80 V. Energy consumption of essential oil by using OHHD at 60, 70, 80 V and HD are shown in Figure 5. The lesser energy consumption of essential oil was by using OHHD at 60, 70 and 80 V then HD.

The change in electrical conductivity of water and Eucalyptus leaves mixture with extraction temperature and extraction time during ohmic heating at different voltage (OHHD at 60, 70, and 80 V) are given in figures 6 and 7. The temperature increased the electrical conductivity of the Eucalyptus leaves increased up to critical temperature of 100 °C and then rate decreased,

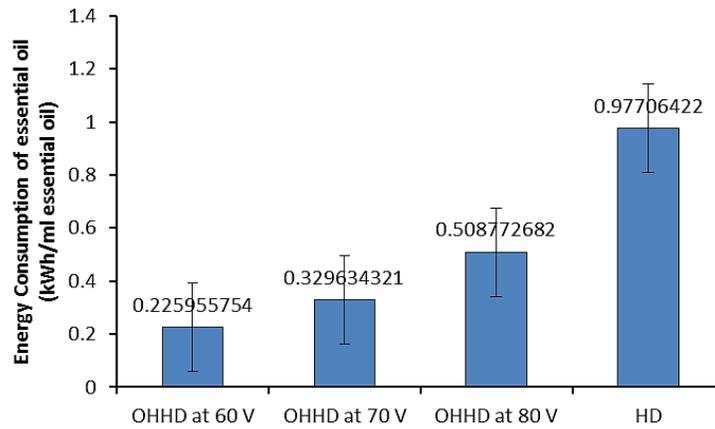


Figure 5. Energy consumption of essential oil at different extraction methods for Eucalyptus leaves.

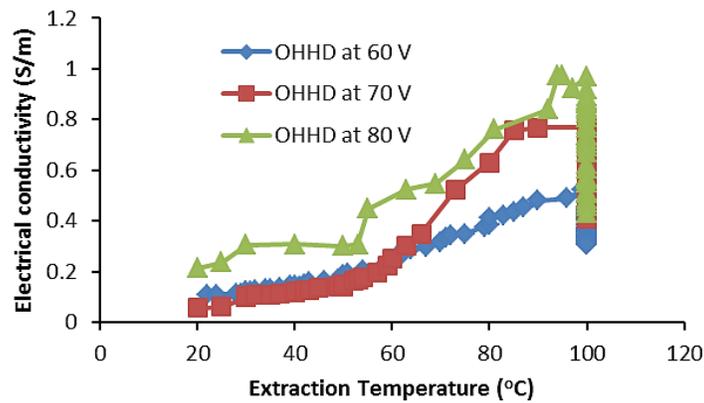


Figure 6. Electric conductivity vs. extraction temperature at different extraction methods for Eucalyptus leaves.

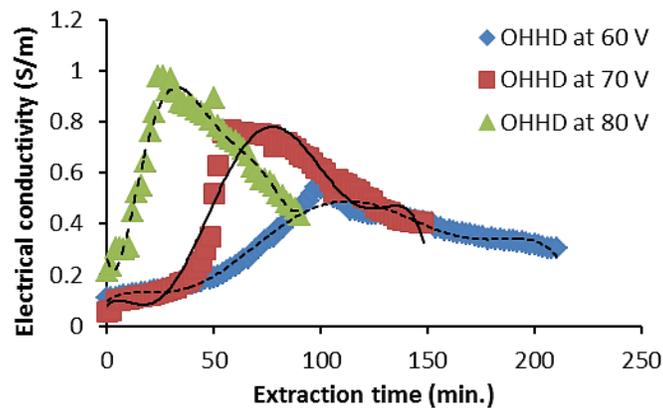


Figure 7. Electric conductivity vs. extraction time at different extraction methods for Eucalyptus leaves.

for all different voltages. When biological tissue is heated, it's electrical conductivity increase due to increase in the ionic mobility (Darvishi et al., 2012).The increasing in the electrical conductivity with temperature is caused by to

the increasing of current with increasing temperature, as Kong et al. (2008), whom stated that the current increase with increasing temperature. Also, the electrical conductivity by using OHHD at 80 V was higher than OHHD

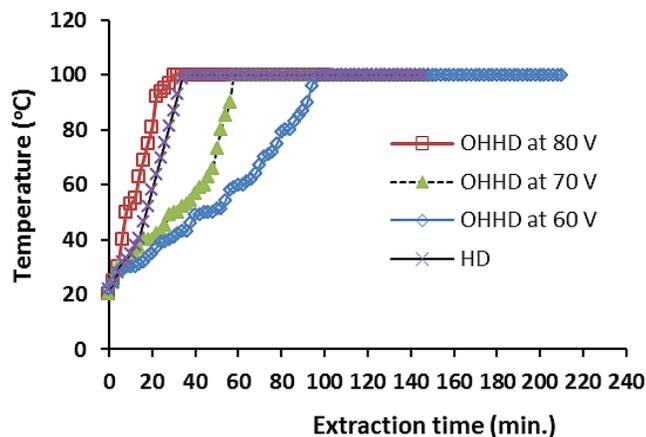


Figure 8. Temperature of essential oil extraction vs. time at different extraction methods for Eucalypts leaves.

Table 2. The heating rate for different extraction methods.

Extraction methods	Heating rate (°C/min.)
OHHD (60 V)	1.157437 ^d
OHHD (70 V)	1.624667 ^c
OHHD (80 V)	2.938113 ^a
HD	1.900997 ^b

OHHD:ohmic heated hydro-distillation,
HD: hydro-distillation

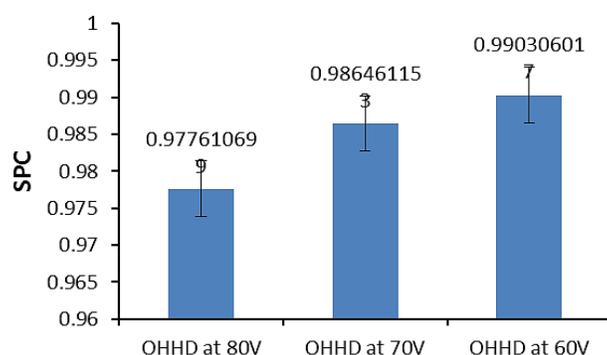


Figure 9. SPC for different extraction methods for Eucalyptus leaves.

at 70 and OHHD at 60 V. The effect of voltage on the ohmic heating times were found to be statistically significant ($P < 0.05$).

As the voltage increased the heating times to reach the prescribed temperature decreased with increasing electrical conductivity, as shown in figure 7.

The temperature during ohmic heated hydro-distillation (OHHD) at 60, 70, 80 and hydrodistillation (HD) extraction are shown in Figure 8. To reach extraction temperature (about 100 °C) and thus obtain the

evaporation of the first essential oil droplets with steam, it was necessary to heat the mix for 30 min. in OHHD at 80 V, while it was 36, 98 and 58 min. for HD, OHHD at 60 and 70 respectively. When current flow through the tissue, the ohmic heating develops. It causes a temperature rise. Table 2 illustrate that heating rate in OHHD (80 V) greater than those of HD and OHHD at 60 and 70 V. These data also indicated that there were significant ($p < 0.05$) differences among HD and OHHD at 60, 70 and 80 V) in terms of heating rate.

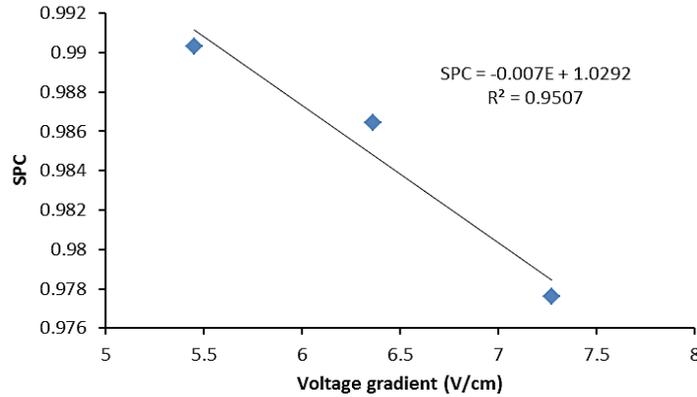


Figure 10.SPC vs voltage gradient at different extraction methods for Eucalyptus leaves.

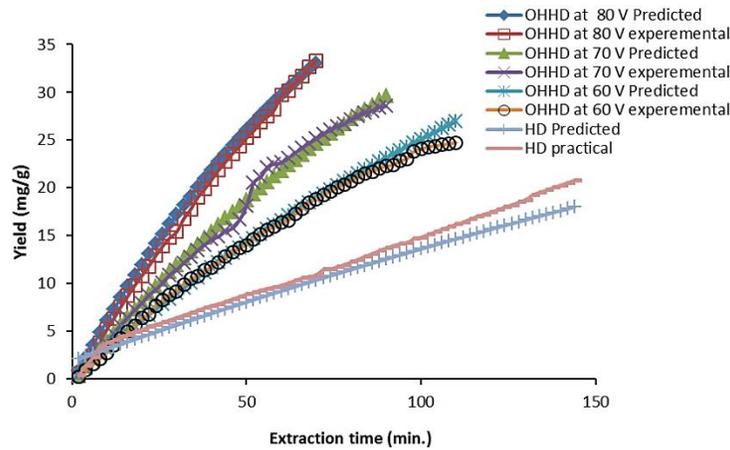


Figure 11. Predicted and experimental yield of essential oil at various distillation methods.

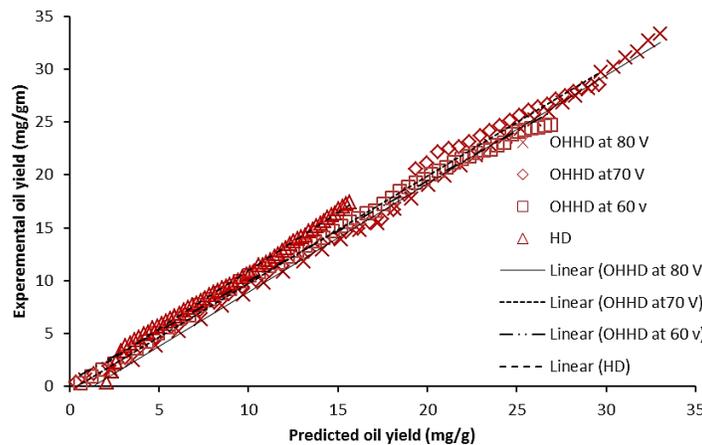


Figure 12. The relationship between the predicted oil yield and experimental at different extraction methods for Eucalypts leaves.

The system performance coefficient (SPC) for various distillation methods (OHHD at 60, 70, 80 V) is shown in figure 9. The SPCs increased from 0.977 to 0.990 as the

voltage decreased. This indicated that 2.238 to 9.693% of the electric energy given to the system was not used in heating up the Eucalyptus leaves. However, for the low

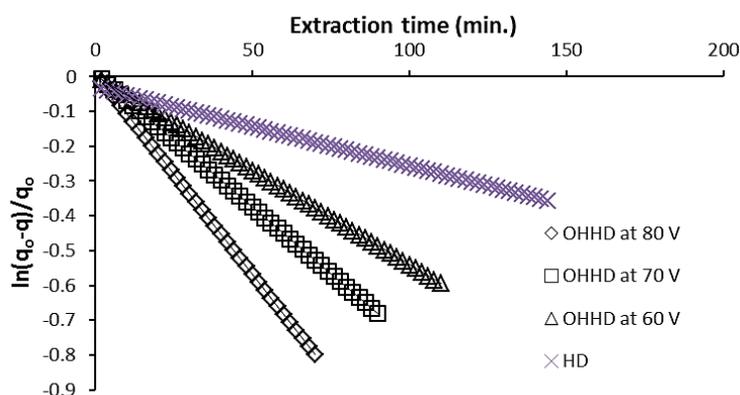


Figure 13. Kinetics of essential oil from experimental data fitted to the mass transfer kinetic model at different distillation methods.

Table 3. The constants and statistical parameters for different extraction methods.

Hydro-distillation methods	A	k (min ⁻¹)	RMSE	RE %	X ²	R ²
OHHD at 60 V	1.00104	0.005422847	0.002949	0.2087952	8.95E-06	0.9982
OHHD at 70 V	1.009489	0.007654434	0.009256	0.7517176	8.82E-05	0.9934
OHHD at 80 V	1.008058	0.011529168	0.011336	1.0161888	0.000132	0.9966
HD	0.969495	0.002260950	0.009191	0.7104643	8.7E-05	0.9927

voltage the conversion of electrical energy into heat was large. Iceier (2003), reported that the SPC values for the liquid samples were in the range of 0.47 to 0.92 during ohmic heating. Darvishi *et al.*, (2012), stated that the SPC for tomato samples were in the range of 0.716 – 0.905.

The relationship between SPCs and voltage gradient was linear equation ($SPC = -0.007E + 1.0292$, $R^2 = 0.9507$) as evident from figure 10. Darvishi *et al.*, (2012) found that the relationship between SPC and voltage gradient is linear equation.

Figure 11 Shows the predicted and experimental essential oil yield (mg/g) from Eucalyptus leaves at various distillation methods (OHHD at 60, 70, 80 V and HD). The predicted and experimental essential oil yield (mg/g) was significantly ($p < 0.05$) increased with increasing extraction time for all treatments. On the other hand, the higher yield of essential oil was found at using OHHD (80 V), but the lower yield of essential oil was found at using HD. The reason of increasing yield by using OHHD (80 V) to increasing of passing current in the Eucalyptus leaves lead to increasing permeability. In the cell membrane, hydrophobic pores are formed by spontaneous thermal fluctuations of membrane lipids. In the cell exposed to an external electric field, the presence of an induced trans membrane provides the free energy necessary for structural rearrangements of membrane

phospholipids and thus enables hydrophilic pore formation (Neumann *et al.*, 1989; Tson, 1991; Chang *et al.*, 1992; Vorobiev and Lebovka, 2008). The result also, illustrates that the shorter total extraction time in OHHD at 80 V was due to the ability of OHHD at 80 V to start the extraction quicker and also because the rate of extraction was greater. The mechanism consisted of two stages: fast oil distillation and slow oil distillation (Pornpunyapat *et al.*, 2011). The oil yield increased rapidly during the initial stage because the oil was rapidly washed from the external surfaces of the Eucalyptus leaves during short period of hydro-distillation. This is called fast oil distillation stage. In the slow oil distillation, the extraction rate gradually slowed down until nearly constant as a result of diffusion of oil from interior of the Eucalyptus leaves towards its surface.

The results also showed that the correlation between the predicted and experimental oil yield was significantly ($p < 0.05$), as well as the coefficient of determination R^2 ranged from 0.9927 - 0.9982. As shown in Table 3. Figure 12 shows the relationship between the predicted and experimental oil yield, all the relationships are linear equations for all hydro-distillation methods.

The dependence of $\ln[(q_0 - q)/q_0]$ versus extraction time is shown in figure 13. It can be seen that equation (11) described experimental data quite well. This data confirm

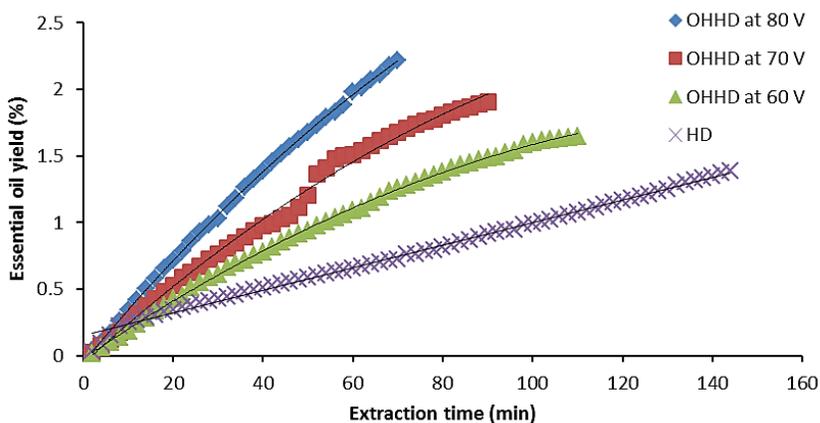
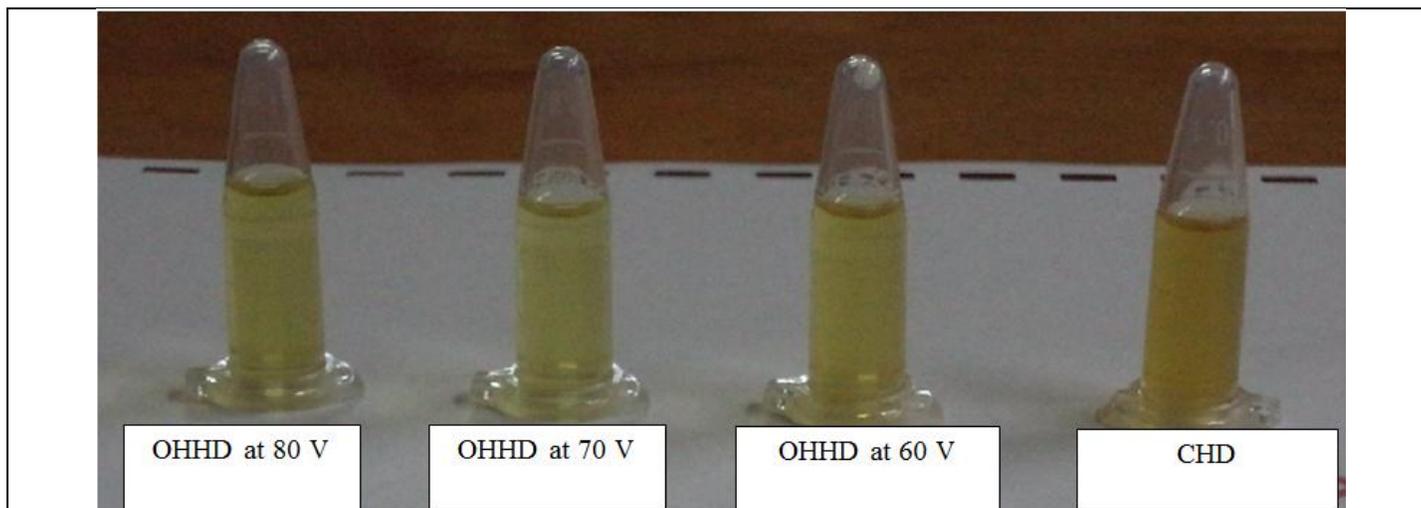


Figure 14. Essential oil yield vs. extraction time at different extraction methods for Eucalyptus leaves.

Table 4. Effect of hydro-distillation methods on the reflective index and density of

Hydro-distillation methods	Specific Gravity	Reflective Index
OHHD 60 V	0.914 ^a ±0.0022	1.4959 ^a ±0.0012
OHHD 70 V	0.913 ^a ±0.0041	1.4945 ^a ±0.0011
OHHD 80 V	0.913 ^a ±0.0033	1.4946 ^a ±0.0014
HD	0.914 ^a ±0.0051	1.4998 ^a ±0.0016

Figure 15. Photograph of essential oils extracted from Eucalyptus leaves by using various distillation methods (OHHD at 60, 70, 80 V and HD).



that the hydro-distillation of Eucalyptus leaves oil was controlled by diffusion stage. Table 3. Illustrate the constants of equation (8) and the statistic parameters for OHHD at 60, 70, 80 V and HD.

The effect of extraction time on essential oil yield is shown in Figure 14, which showed that the essential oil yield (%) was increased with increasing extraction time for OHHD at 60, 70, 80 V and HD. In addition, the

relationship between essential oil yield and extraction time was second order equation except HD was linear Equation.

The needed time to essential oil extraction was 110, 90, 70 and 142 min for OHHD at 60, 70, 80 V and HD respectively. This is due to the more efficient in the ohmic heated hydro-distillation. Ohmic heating can heat the entire heating rate, therefore, it's able to generate heat

Figure 16. GC-Mass chromatography of essential oil from Eucalyptus leaves by OHHD at 60 V.

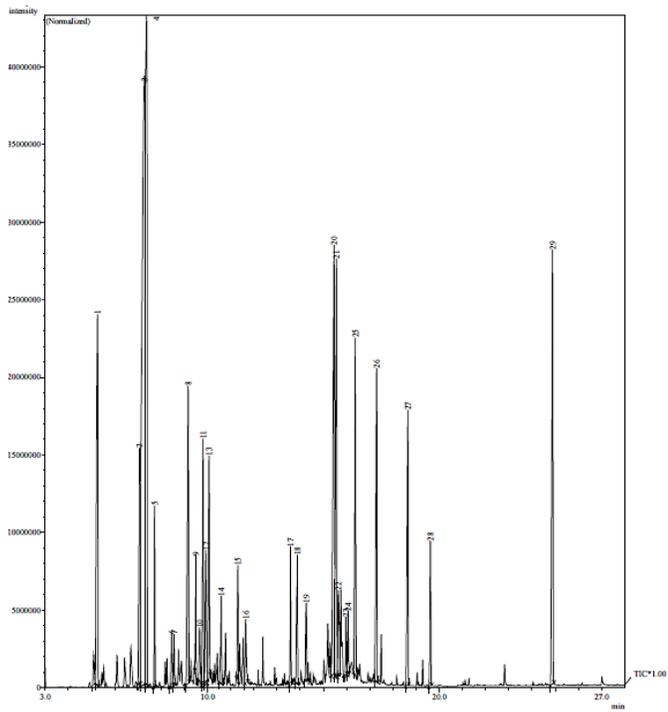


Figure18. GC-Mass chromatography of essential oil from Eucalyptus leaves by OHHD at 80 V.

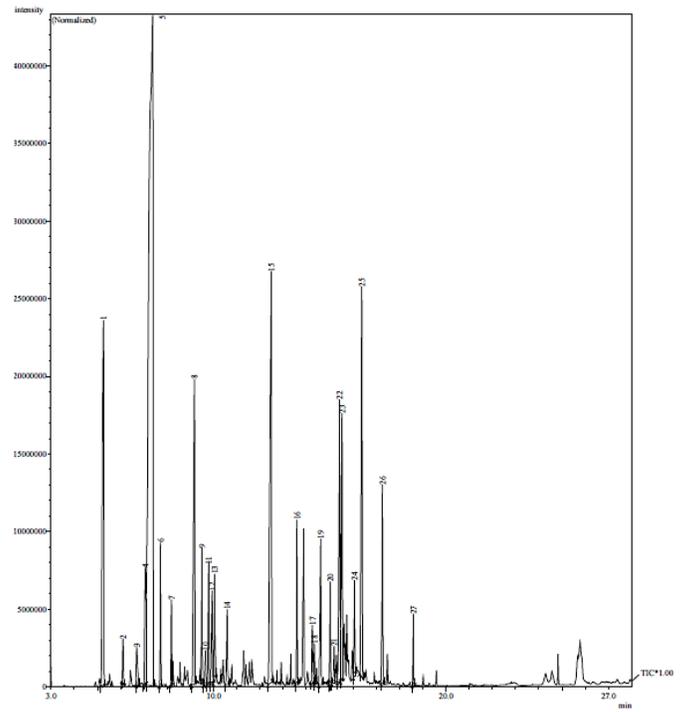


Figure 17. GC-Mass chromatography of essential oil from Eucalyptus leaves by OHHD at 70 V.

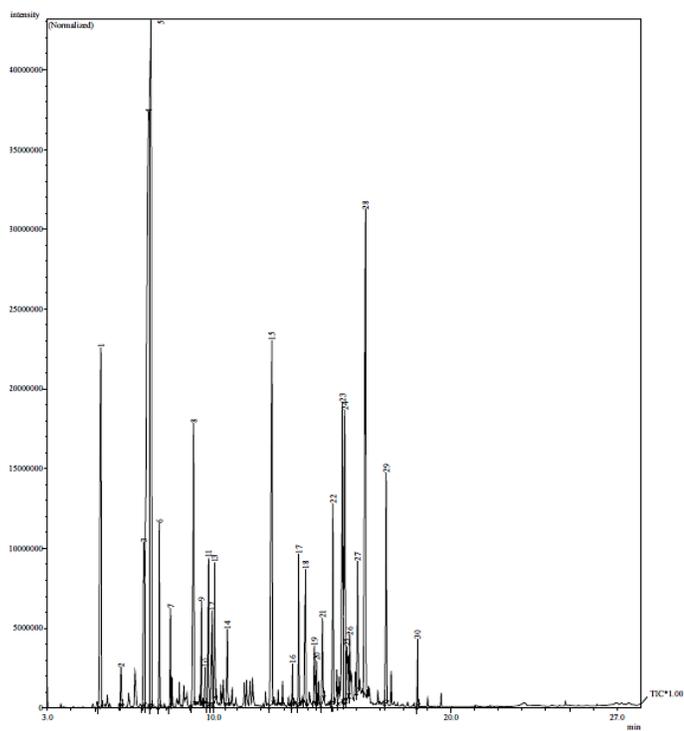


Figure19. GC-Mass chromatography of essential oil from Eucalyptus leaves by HD.

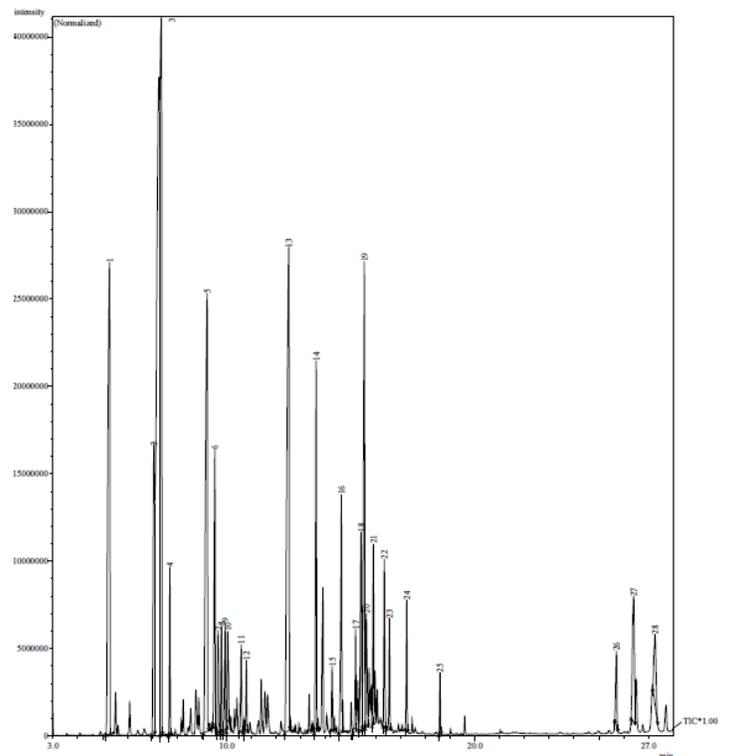


Table 5. Chemical compounds of Eucalyptus oil which extracted by OHHD at 60 V.

No.	Chemical compound	R.T (min)	Formula	Area (%)	Mol. Weight	Height (%)
1	alpha.-Pinene	5.255	C10H16	6.07	136	5.79
2	Benzene, 1-methyl-3-(1-methylethyl)-	7.067	C10H14	3.67	134	3.69
3	Benzene, 1-methyl-3-(1-methylethyl)-	7.267	C10H14	22.09	134	9.41
4	Eucalyptol	7.373	C10H18O	12.54	154	10.45
5	1,4-Cyclohexadiene, 1-methyl-4-(1-methyle)	7.716	C10H16	1.42	136	2.82
6	1,6-Octadien-3-ol, 3,7dimethyl-	8.454	C10H18O	0.69	154	0.82
7	Butanoic acid, 3-methyl-, 3-methylbutyl este	8.559	C10H20O2	0.45	172	0.80
8	Bicyclo[3.1.1]heptan-3-ol, 6,6-dimethyl-2-m	9.168	C10H16O	4.11	152	4.64
9	2(10)-Pinen-3-one, (+/-)-	9.495	C10H14O	1.00	150	1.88
10	Borneol	9.660	C10H18O	0.68	154	0.84
11	3-Cyclohexen-1-ol, 4-methyl-1-(1-methyleth	9.809	C10H18O	2.68	154	3.82
12	Bicyclo[3.2.0]heptan-3-ol, 2-methylene-6,6-	9.922	C10H16O	1.88	152	2.08
13	3-Cyclohexene-1-methanol, .alpha.,.alpha.,4	10.067	C10H18O	2.59	154	3.47
14	Cyclohexanol, 2-methylene-5-(1-methylethe	10.591	C10H16O	1.00	152	1.37
15	1-Cyclohexene-1-carboxaldehyde, 4-(1-meth	11.311	C10H16O	0.95	152	1.80
16	Phenol, 2-methyl-5-(1-methylethyl)-	11.649	C10H14O	0.80	150	0.99
17	1H-Cycloprop[e]azulene, decahydro-1,1,7-t	13.585	C15H24	1.17	204	2.16
18	1H-Cycloprop[e]azulene, decahydro-1,1,7-tr	13.875	C15H24	1.58	204	2.01
19	Oxalic acid, monoamide, N-(2-phenylethyl)	14.257	C16H23NO3	0.91	277	1.21
20	1H-Cycloprop[e]azulen-7-ol, decahydro-1,1	15.458	C15H24O	7.39	220	6.71
21	(-)-Globulol	15.560	C15H26O	5.66	222	6.51
22	Cubenol	15.644	C15H26O	1.28	222	1.34
23	2-Naphthalenemethanol, 2,3,4,4a,5,6,7,8-oct	15.974	C15H26O	0.45	222	0.91
24	2-Naphthalenemethanol, 1,2,3,4,4a,5,6,7-oc	16.067	C15H26O	0.62	222	0.99
25	2-Naphthalenemethanol, decahydro-.alpha.,.	16.375	C15H26O	3.85	222	5.27
26	Spiro[5.5]undec-2-ene, 3,7,7-trimethyl-11-m	17.296	C15H24	3.96	204	4.89
27	1,2-Benzenedicarboxylic acid, bis(2-methyl	18.640	C16H22O4	3.34	278	4.29
28	1,2-Benzenedicarboxylic acid, butyl 2-methy	19.619	C24H38O4	1.37	390	2.26
29	1,2-Benzenedicarboxylic acid, diisooctylest	24.889		5.80		6.77

side product. (Goullieux and Pain, 2005; Sastry,2005). Essential oil yield (%) by using OHHD at 60, 70, 80 V and HD were 1.648, 1.894, 2.177 and 1.369 % respectively.

Results showed that using OHHD at 80 V was gave higher essential oil yield compared with other treatments. The values of reflective index and density of the extracted Eucalyptus oil by using various distillation methods (OHHD at 60, 70, 80 V and HD) are given in table 4. There is no significant differences among OHHD at 60,

70, 80 V and HD for the specific gravity and reflective index. Alhakeem and Hasan (1985) stated that the reflective index and specific Gravity for extracted Eucalyptus oil are ranged between 1.4631 – 1.4644 and 0.9160 -0.9300 respectively. Abdull-Neby (2009), found that the reflective index and specific gravity for extracted Eucalyptus oil by using hydro-distillation method are 1.4928 and 0.9162 respectively. Therefore, taking into account physical properties (specific gravity, reflective

Table 6. Chemical compounds of Eucalyptus oil extracted by OHHD at 70 V.

No.	Chemical compound	R.T (min)	Formula	Area (%)	Mol. Weight	Height (%)
1	alpha.-Pinene	5.247	C10H16	6.37	136	6.32
2	Bicyclo[3.1.1]heptane, 6,6-dimethyl-2-meth	6.096	C10H16	0.44	136	0.66
3	Benzene, 1-methyl-3-(1-methylethyl)-	7.056	C10H14	2.68	134	2.88
4	Benzene, 1-methyl-3-(1-methylethyl)-	7.258		21.65		10.43
5	Eucalyptol	7.363	C10H18O	15.66	154	12.08
6	1,4-Cyclohexadiene, 1-methyl-4-(1-methyle	7.715	C10H16	1.69	136	3.24
7	(+)-4-Carene	8.181	C10H16	0.88	136	1.73
8	Bicyclo[3.1.1]heptan-3-ol, 6,6-dimethyl-2-	9.160	C10H16O	4.25	152	4.95
9	2(10)-Pinen-3-one, (+/-)-	9.486	C10H14O	0.95	150	1.79
10	Bicyclo[2.2.1]heptan-2-ol, 1,7,7-trimethyl-	9.642	C10H18O	0.41	154	0.61
11	3-Cyclohexen-1-ol, 4-methyl-1-(1-methylet	9.786	C10H18O	1.92	154	2.59
12	Cyclohexanol, 2-methylene-5-(1-methylethe	9.930	C10H16O	1.32	152	1.67
13	3-Cyclohexene-1-methanol, .alpha.,.alpha.,4	10.044	C10H18O	1.62	154	2.45
14	Cyclohexanol, 2-methylene-5-(1-methylethe	10.578	C10H16O	0.91	152	1.32
15	3-Allyl-6-methoxyphenol	12.458	C10H12O2	6.53	164	6.41
16	Caryophyllene	13.323	C15H24	0.37	204	0.74
17	1H-Cycloprop[e]azulene, decahydro-1,1,7-t	13.586	C15H24	1.46	204	2.66
18	1H-Cycloprop[e]azulene, decahydro-1,1,7-t	13.877	C15H24O	2.02	220	2.37
19	.beta.-Guaiene	14.248	C15H24	0.69	204	1.00
20	2-Isopropenyl-4a,8-dimethyl-1,2,3,4,4a,5,6,	14.342	C15H24	0.40	204	0.76
21	Phenol, 2-methoxy-4-(2-propenyl)-, acetate	14.591	C12H14O3	0.98	206	1.47
22	Cyclohexanemethanol, 4-ethenyl-.alpha.,.al	15.033	C15H26O	2.36	222	3.53
23	1H-Cycloprop[e]azulen-7-ol, decahydro-1,1	15.430	C15H24O	4.83	220	5.28
24	(-)-Globulol	15.530	C15H26O	4.50	222	5.13
25	(-)-Globulol	15.620	C15H26O	0.92	222	0.93
26	Ledol	15.737	C15H26O	0.49	222	0.91
27	2-Naphthalenemethanol, 1,2,3,4,4a,5,6,7-oct	16.071	C15H26O	1.65	222	2.30
28	2-Naphthalenemethanol, decahydro-.alpha.,.	16.404	C15H26O	8.69	222	8.54
29	.gamma.-Neoclovene	17.277	C15H24	2.76	204	4.06
30	1,2-Benzenedicarboxylic acid, bis(2-methyl	18.597	C16H22O4	0.61	278	1.19

index and color) of the extracted essential oil from Eucalyptus leaves using OHHD at 60, 70, 80V as a new extraction technique, did not cause any problems to the essential oil extracted from Eucalyptus leaves. On the other hand, Figure 15. Illustrate photograph of essential oils extracted from Eucalyptus leaves by using various distillation methods (OHHD at 60, 70, 80 V and HD). The differences is that the color of the essential oils extracted by OHHD at 70 and 80 V is lighter than that obtained by HD and OHHD at 60 V.

Chemical Composition of the Essential Oil

Figures from 16 to 19 and tables from 5 to 8 show the component of essential oil of Eucalyptus leaves as analyzed by GC-MS. A total of 29, 30, 27 and 28 components were detected at using OHHD at 60, 70, 80 V and HD respectively, representing 100% of the oil from all treatments. The principle component of Eucalyptus essential oil was Eucalyptol. The mean values of Eucalyptol content of Eucalyptus essential oil of from OHHD at 60, 70,

Table 7. Chemical compounds of Eucalyptus oil extracted by OHHD at 80 V.

No.	Chemical compound	R.T (min)	Formula	Area (%)	Mol.Weight	Height (%)
1	.alpha.-Pinene	5.255	C10H16	7.46	136	7.94
2	Bicyclo[3.1.1]heptane, 6,6-dimethyl-2-meth	6.098	C10H16	0.54	136	0.98
3	.alpha.-Phellandrene	6.689	C10H16	0.70	136	0.83
4	Benzene, 1-methyl-3-(1-methylethyl)-	7.054	C10H14	2.04	134	2.53
5	Eucalyptol	7.379	C10H18O	39.49	154	14.58
6	1,4-Cyclohexadiene, 1-methyl-4-(1-methyle	7.714	C10H16	1.42	136	3.12
7	(+)-4-Carene	8.182	C10H16	0.80	136	1.87
8	Bicyclo[3.1.1]heptan-3-ol, 6,6-dimethyl-2-	9.173	C10H16O	5.27	152	6.63
9	2(10)-Pinen-3-one, (+/-)-	9.497	C10H14O	1.39	150	2.90
10	Isoborneol	9.646	C10H18O	0.53	154	0.75
11	3-Cyclohexen-1-ol, 4-methyl-1-(1-methylet	9.788	C10H18O	1.60	154	2.61
12	Cyclohexanol, 2-methylene-5-(1-methylethe	9.931	C10H16O	1.42	152	2.03
13	3-Cyclohexene-1-methanol, .alpha.,.alpha.,4	10.042	C10H18O	1.35	154	2.32
14	Cyclohexanol, 2-methylene-5-(1-methylethe	10.581	C10H16O	1.04	152	1.64
15	3-Allyl-6-methoxyphenol	12.481	C10H12O2	9.46	164	8.94
16	1H-Cycloprop[e]azulene, decahydro-1,1,7-t	13.589	C15H24	1.68	204	3.58
17	1H-Benzocycloheptene, 2,4a,5,6,7,8-hexahy	14.249	C15H24	0.79	204	1.26
18	1,5-Cyclodecadiene, 1,5-dimethyl-8-(1-met	14.342	C15H24	0.41	204	0.87
19	Phenol, 2-methoxy-4-(2-propenyl)-, acetate	14.607	C12H14O3	2.00	206	3.18
20	Cyclohexanemethanol, 4-ethenyl-.alpha.,.al	15.022	C15H26O	1.19	222	2.22
21	Epiglobulol	15.192	C15H26O	0.53	222	0.80
22	1H-Cycloprop[e]azulen-7-ol, decahydro-1,1	15.425	C15H24O	4.50	220	6.14
23	(-)-Globulol	15.530	C15H26O	4.14	222	5.83
24	2-Naphthalenemethanol, 1,2,3,4,4a,5,6,7-oct	16.066	C15H26O	1.19	222	2.09
25	2-Naphthalenemethanol, decahydro-.alpha.,.	16.381	C15H26O	5.83	222	8.49
26	.gamma.-Neoclovene	17.272	C15H24	2.52	204	4.33
27	1,2-Benzenedicarboxylic acid, bis(2-methyl	18.598	C16H22O4	0.71	278	1.56

80 V and HD were 12.54, 15.66, 39.49 and 15.96 % respectively. Other major components of Eucalyptus essential oil from OHHD at 60, 70, 80 V and HD area were {alpha-Pinene (6.07%), Benzene,1-methyl-3-(1-methylethyl)-(22.09%), 1H-Cycloprop[e]azulen-7-ol,decahydro-1,1(7.39%) and (-)-Globulol (5.66%)}, {alpha-Pinene (6.37%), Benzene, 1-methyl-3-(1-methylethyl)-(21.65%),3-Allyl-6-methoxyphenol(6.53%), 2-Naphthalenemethanol,decahydro-.alpha.,(8.69%)}, {alpha-Pinene(7.46), Bicyclo[3.1.1]heptan-3-ol, 6,6-dimethyl-2-(5.27%), 3-Allyl-6-methoxyphenol(9.46%), 2- Naphthalenemethanol, decahydro-.alpha.,(5.83)}, {alpha-Pinene(11.40%), Benzene, 1-methyl-3-(1-methylethyl)-

(5.16%), Bicyclo[3.1.1]heptan-3-ol, 6,6-dimethyl-2-(10.25%), 3-Allyl-6-methoxyphenol(12.47%), (-)-Globulol (6.85%)} respectively. The results showed that the Eucalyptol content was strongly affected by ohmic heating. In the OHHD at 60, 70, 80 V and HD, the oil consists of 72.41, 56.66, 62.96 and 85.71% respectively Oxygen at edmonoterpenes compounds and 27.58, 43.33, 37.03 and 14.28 % monoterpenes hydrocarbons. The results agreed with Iqbal et al., (2006) were found that Eucalyptus oil components contain of 17.46% Eucalyptol and 3.35% alpha-Pinene. Compounds Eucalyptus oil (*E.robusta*) are 73% alpha-Pinene, 1.6% Terpinolene (Sartorelli et al., 2007).

Table 8. Chemical compounds of Eucalyptus oil extracted by HD.

No.	Chemical compound	R.T (min)	Formula	Area (%)	Mol. Weight	Height (%)
1	alpha.-Pinene	5.279	C10H16	11.40	136	8.01
2	Benzene, 1-methyl-3-(1-methylethyl)-	7.063	C10H14	5.16	134	4.89
3	Eucalyptol	7.364	C10H18O	15.96	154	12.16
4	1,4-Cyclohexadiene, 1-methyl-4-(1-methyle)	7.708	C10H16	1.46	136	2.85
5	Bicyclo[3.1.1]heptan-3-ol, 6,6-dimethyl-2-	9.213	C10H16O	10.25	152	7.43
6	2(10)-Pinen-3-one, (.+/-)-	9.522	C10H14O	3.42	150	4.74
7	Bicyclo[2.2.1]heptan-2-ol, 1,7,7-trimethyl-	9.657	C10H18O	1.08	154	1.73
8	3-Cyclohexen-1-ol, 4-methyl-1-(1-methylet	9.787	C10H18O	1.21	154	1.80
9	Cyclohexanol, 2-methylene-5-(1-methylethe	9.934	C10H16O	1.38	152	1.85
10	3-Cyclohexene-1-methanol, .alpha.,.alpha.,4	10.042	C10H18O	1.35	154	1.63
11	Cyclohexanol, 2-methylene-5-(1-methylethe	10.584	C10H16O	1.09	152	1.48
12	2-Cyclohexen-1-one, 2-methyl-5-(1-methyle	10.792	C10H14O	0.83	150	1.22
13	3-Allyl-6-methoxyphenol	12.504	C10H12O2	12.47	164	8.18
14	1H-Cycloprop[e]azulene, decahydro-1,1,7-t	13.607	C15H24	4.08	204	6.29
15	3-Phenylpropanoic acid, dodec-9-ynyl ester	14.249	C21H30O2	0.70	314	1.09
16	Phenol, 2-methoxy-4-(2-propenyl)-, acetate	14.624	C12H14O3	2.97	206	4.04
17	Epiglobulol	15.198	C15H26O	1.16	222	1.74
18	1H-Cycloprop[e]azulen-7-ol, decahydro-1,1	15.418	C15H24O	3.15	220	3.35
19	(-)-Globulol	15.548	C15H26O	6.85	222	7.91
20	Cubenol	15.632	C15H26O	1.54	222	1.93
21	Apiol	15.913	C12H14O4	1.89	222	3.06
22	2-Naphthalenemethanol, decahydro-.alpha.,.	16.353	C15H26O	1.90	222	2.85
23	Apiol	16.559	C12H14O4	1.08	222	1.90
24	4,6,6-Trimethyl-2-(3-methylbuta-1,3-dienyl	17.256	C15H22O	1.27	218	2.22
25	1,2-Benzenedicarboxylic acid, bis(2-methyl	18.597	C16H22O4	0.54	278	1.05
26	12-Oleanen-3-yl acetate, (3.alpha.)-	25.709	C32H52O2	1.05	468	1.27
27	4,4,6a,6b,8a,11,11,14b-Octamethyl-1,4,4a,5	26.405	C30H48O	3.07	424	2.13
28	9,19-Cyclolanost-24-en-3-ol, (3.beta.)-	27.268	C30H50O	1.67	426	1.20

CONCLUSIONS

In conclusion, OHHD at 80 V was better than OHHD at 60, 70 V and HD of oil yield (%) and shorter extraction time. The proposed second order kinetic model fitted well with the experimental data. OHHD resulted in a reduced extraction time and an energy saving compared to the HD. Energy consumption, current and input power using OHHD were little than HD. On the other hand, electrical conductivity was increased with increasing temperature. Heating rate by using OHHD at 80 V was higher than

OHHD at 60, 70 V and HD. SPC reduced with increasing voltage gradient and its value ranged between 0.977 – 0.990. The determination coefficient between predicted oil yield and experimental ranged between 0.9927 to 0.9982. The results showed, there is no significant differences among OHHD at 60, 70, 80 V and HD for the specific gravity and reflective index. The principle component of Eucalyptus essential oil was Eucalyptol. The mean values of Eucalyptol content of Eucalyptus essential oil of from OHHD at 60, 70, 80 V and HD were 12.54, 15.66, 39.49 and 15.96 % respectively.

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