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EFFECT OF DISTILATION TIME AND STORAGE PERIOD ON THE PHYSICOCHEMICAL PROPERTIES OF LEMONGRASS OIL

BY

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ABSTRACT

The yield of lemongrass volatile oil, as affected by time of distillation was 0.20%, 0.28%, 0.28%, 0.28% and 0.28% after one, two, three, four and five hours, respectively. The oil yield after two hours of distillation was higher than after one hour and remained stable after that. Physicochemical properties of the oil samples were determined in each case. The oils were analysed by gas chromatography to identify the different components in the samples. The values of aldehyde content were calculated as citral and the main component of the oil after 1, 2, 3, 4 and 5 hours of distillation were 58.32%, 61.42%, 60.44%, 59.21% and 57.83%, respectively. The gas chromatography analysis proved that the chemical composition of each oil was different. Citral was the main component of the oil which amounted to 37.18%, 36.57%, 33.75%, 31.32% and 30.20% after 1, 2, 3, 4 and 5 hours, respectively. Two hours of distillation may be recommended to obtain the oil of lemongrass which gave the high yield of oil besides to its high citral content. Storage of essential oils is considered as one of the most important factors which affect the physicochemical properties of these oils as well as their rate of deterioration. The specific gravity, refractive index, acid number, ester number after acetylation and aldehydes increased gradually during long-term storage. Oil stored at 5±2°C showed slower rates of changes in most physicochemical properties during storage.

By gas chromatographic analysis, 10 components were identified. These components namely: α-pinene, limonene, linalool, geraniol, thymol, linaly acetate, geranyl acetate, citral, methene and anicic.

INTRODUCTION

The lemongrass plant grows wild in many tropical and semitropical parts of Asia, Africa and America. The trade distinguishes between two principal types of lemongrass oil, the East Indian and the so called West Indian oil. Both oils contain from 75 to 85% aldehydes, but they differ in their solubility in 70% ethyl alcohol. The increasing interest regarding the use of aromatic plants and their products as a possible source of national income, especially hard currency, must

be accompanied with an extensive amount of research to investigate both the agricultural requirements of such plants and the subsequent process leading to the production of essential oils of high quality to meet the requirements specified by those interested in such products and their industrial applications.

Lemongrass oil which is widely used industrially and medicinally. The value of lemongrass oil depends almost entirely upon its citral content which is generally utilized industrial and medicinal purposes. Guenther (1961) mentioned that the oil of lemongrass has become one of the most important and indispensable essential oils and large quantities of this oil are now employed for the manufacture of synthetic vitamin A. Keimi *et al.* (1953) indicated that lemongrass oil has an effect on the inhibition of *Micrococcus pyogenes* var. aureus and *Mycobacterium tuberculosis*. Umney (1913), reported the use of lemongrass oil as a good source of income, from this oil extensive medical claims are made and it is used as a household remedy for the cure of almost every complaint. Guenther (1961), referred to the conversion of the spent leaf material after distillation into paper, while Noemi (1939) noted that lemongrass plants is a good crop to minimize soil erosion. Paul and Bedoukian (1950) added that citral has extensive application as a flavoring and perfumery ingredient. Because of its lemon-like odour, oil of lemongrass is used widely for scenting soaps, cosmetics, synthetic lemon, lime, orange flavours and all kinds of technical products.

Tawfik (1980), mentioned that the citral content was 75% in lemongrass oil. El-Zahwey *et al.* (1992) identified the following components of the lemongrass oil: camphene, β-pinene, limonene, linalyl acetate, geranyl acetate, geraniol citral and citronellal.

MATERIALS AND METHODS

Materials:

Lemongrass oil was obtained by steam distillation of lemongrass plants grown in the research grounds of the Medicinal and Aromatic Plants Section of the Ministry of Agriculture at Barrage, A.R.E. The lemongrass oil used in this investigation was obtained by steam distillation according to Guenther (1961). By steam distillation method the lemongrass oil samples were obtained after 1, 2, 3, 4 and 5 hours of distillation. Oil samples were dried over pure anhydrous sodium sulphate (120-150 g/kg of oil). The mixture was first allowed to stand at room temperature for 24 hours and then filtered through ordinary filter paper. The obtained oil samples were kept in washed, cleaned and dried brown glass bottles which were completely filled by the oil. The bottles were stoppered carefully and were tightly sealed by molten wax. The bottles were then stored at 5°C in order to preserve the physical and chemical properties.

Methods:

The essential oil content of lemongrass was determined according to the British Pharmacopoeia Codex Method (1963).

The effect of temperature on the physical and chemical properties of lemongrass oil was studied.

Prown bottles, completely filled and stored at room temperature (15-30°C depending on the season), and low temperature at 5±2°C. The physicochemical properties of tested oil (specific gravity, refractive index, optical rotation, acid number, ester number and ester number after acetylation) were determined according to the methods described by Guenther (1961).

The gas chromatographic technique was used to separate and identify the components of lemongrass oil. The conditions used are described in these table.

Analysis conditions of gas liquid chromatography of lemongrass oil.

Item	Condition
Instrument type	G.C.V. pye unicum
Column packing	PEGA 10%
Flow rate	H_2: 33 ml/min. N_2: 30 ml/min. Air: 33 ml/min.
Column temperature	60°C. progm. 4°C/min. up to 180°C
Injector temperature	300°C
Detector temperature	220°C
Chart speed	0.5 cm/min.
Sample size	1 µl

RESULTS AND DISCUSSION

The essential oil percent obtained by steam distillation from lemongrass plants was 0.2%, 0.28%, 0.28%, 0.28% and 0.28% after 1, 2, 3, 4 and 5 hours, respectively. This result showed that the oil percent after 2 hours of distillation was higher than that obtained after one hour and became stable after that. Therefore it could be said that 2 hours distillation may be quit sufficient for obtaining most of the essential oil from lemongrass plants, however Guenther (1961) reported that, distillation of lemongrass plant, lasts about three hours to produce the oil.

Lemongrass oil samples were analysed for their specific gravity, refractive index, optical rotation, solubility, acid number, ester number, ester number after acetylation and aldehydes. The obtained results are shown in Table (1) and (2).

From Table (1) it could be concluded that the specific gravity of the lemongrass oil samples obtained after 1, 2, 3, 4 and 5 hours of distillation was 0.8832, 0.8841, 0.8865, 0.8866 and 0.8866, respectively. It could be observed that the specific gravity of the oil samples obtained after 2, 3, 4 and 5 hours of distillation was higher than that obtained after the first hour. The specific gravity of oil samples slightly increased by increasing time of distillation.

Table (1): Effect of distillation time on the physical properties of lemongress oil.

Distillation time (hr)	Physical properties		
	Specific gravity at 15°C	Refractive index at 20°C	Optical rotation
1	0.8832	1.4801	-0.28°
2	0.8841	1.4812	-0.56°
3	0.8865	1.4815	-0.78°
4	0.8866	1.4818	-1.07°
5	0.8866	1.4818	-1.44°

* Oil obtained after 1 hr of distillation was clearly soluble in 1.0-1.2 vol. of 80% alcohol and more up to 10 vols.
* Oil obtained after 2 hrs of distillation was clearly soluble in 1.0-1.2 vol. of 90% alcohol and more up to 10 vols.
* Oil obtained after 3 hrs of distillation was clearly soluble in 0.6-2.0 vol. of 80% alcohol and more up to 10 vols.
* Oil obtained after 4 hrs of distillation was clearly soluble in 0.7-1.1 vol. of 80% alcohol and more up to 10 vols.
* Oil obtained after 5 hrs of distillation was clearly soluble in 0.6-1.0 vol. of 80% alcohol and more up to 10 vols.

Table (2): Effect of distillation time on Chemical properties of lemongrass oil.

Distillation time (hr)	Acid number	Ester number	Ester content calculated as linalyl acetate	Ester number after acetylation	Aldehyde content calculated as citral %
1	2.55	12.08	5.69	201.73	58.32
2	3.08	16.57	7.81	201.62	60.24
3	3.27	16.87	7.95	198.49	61.44
4	3.69	20.43	9.62	193.23	59.21
5	3.75	24.47	11.53	190.54	57.83

The refractive index at 20°C took the same trend and increased slightly by increasing time of distillation as it was 1.4801, 1.4812, 1.4815, 1.4818 and 1.4818, respectively. These results are in agreement with this reported by Soad (1973).

The optical rotation of lemongrass oil samples was determined at room temperature. The optical rotation of the oil samples obtained after 1, 2, 3, 4 and 5 hours of distillation were -0.28°, -0.56°, -0.78°, -1.07° and -1.44°, respectively. The optical rotation after 1 hour of distillation was lower in comparison with the other oil samples obtained after 2, 3, 4 and 5 hours. These results are in agreement with those mentioned by El-Zahwey (1978).

Concerning solubility in alcohol all lemongrass essential oils obtained were insoluble in 70% of alcohol, however they were soluble in different vols. of either 80 or 90% of ethyl alcohol. These results agree with values reported by Guenther (1961) and El-Zahwey *et al.* (1992).

The acid number of lemongrass oils increased gradually as it was, 2.55, 3.08, 3.27, 3.69 and 3.75 after 1, 2, 3, 4 and 5 hours of distillation respectively, which might be due to partically oxidation of certain alcohols and aldehydes to acids through long time of distillation. These results are in agreement with those mentioned by Mahmoud (1977) and El-Zahwey *et al.* (1992).

The ester number and ester content calculated as linalyl acetate of oils took the same trend and increased gradually as it was 12.08, 16.57, 16.87, 20.43, 24.47 and 5.69, 7.81, 7.95, 9.62, 11.53 after 1, 2, 3, 4 and 5 hours of distillation, respectively.

On the contrary, the ester number after acetylation of lemongrass oils obtained decreased gradually as it reached 201.73, 201.62, 198.49, 193.23 and 190.54 after 1, 2, 3, 4 and 5 hours of distillation, respectively.

It is well known that aldhydes content in lemongrass essential oils is the most important compound that values its quality. Aldehyde content in oils obtained calculated as citral fluctuated between 57.83 and 61.44 after different hours of distillation.

Sensitive substances as aldhydes tend to polymerise under long time of high temperature as steam distillation which may explain such increase in the specific gravity after 5 hours of distillation of lemongrass essential oil.

Data obtained fell within the range of those mentioned by Rutovski (1970), Soad (1973), Mahmoud *et al.* (1983) and El-Zahwey *et al.* (1992).

Gas liquid chromatography (G.L.C.) analysis of lemongrass oil samples:

The gas chromatography technique was used to separate and identify the main components of lemongrass oil obtained after 1, 2, 3, 4 and 5 hours of distillation and data obtained are shown in Figures 1, 2, 3, 4 and 5, respectively.

The chromatograms showed 16 peaks for lemongrass essential oil sample produced after 1, 2, 3, 4 and 5 hours of distillation, where ten of these peaks were identified. The components of these oils may be classified into 6 categories namely: hydrocarbons, alcohols, esters, aldehydes, ketones and acids.

The hydrocarbons namely α–pinene and limonene, represented the peaks numbers 1 and 2 respectively.

The identified alcohols linalool, geraniol and thymol, represented the peaks numbers 6, 7 and 16 respectively. Geraniol was the highest alcohol which

amounted 28.39%, 30.62%, 24.82%, 23.65% and 29.46% after 1, 2, 3, 4 and 5 hours of distillation respectively.

The esters namely linalyl acetate and geranyl acetate, represented the peaks numbers 7 and 14, respectively. Linalyl acetate was higher than the geranyl acetate which amounted 5.16%, 4.61%, 3.82%, 1.45% and 4.01% after 1, 2, 3, 4 and 5 hours of distillation respectively.

Regarding to aldehydes, citral was the main component identified in the oils which amounted to 37.18%, 36.57%, 33.75%, 31.32% and 30.20% after 1, 2, 3, 4 and 5 hours of distillation respectively, hence decrement of citral content may be due to undergoing oxidizing effect associated with heat during steam distillation. These results with those reported by Guenther (1961).

The ketone component namely menthene and the acid namely anicic, these components represented the peaks No. 5 and 15 respectively.

From the above mentioned results, it could be summarized that steam distillation of lemongrass plants for 2 hours revealed higher quality of essential oil due to its high content of citral as assured by Guenther (1961).

Storage of lemongrass oil:

Essential oils of lemongrass obtained by steam distillation for 1, 2, 3, 4 and 5 hours were stored for 12 months at room and low temperature. Dark bottles filled with these oils were stored at room temperature and the physicochemical properties were investigated every 3 months. Results obtained are shown in Table (3) specific gravity, refractive index, acid number, ester number after acetylation and aldehyde content calculated as citral increased gradually after storage for 360 days, while optical rotation and ester number decreased. These changes might be due to hydrolysis, causing increase in acid number and ester number after acetylation besides decrease in ester number and optical rotation which is probably be as a result of changes in the chemical structure of some compounds in oils, however these oils were considered available for marketing after this long period of storage. The quality of the oil stored at room temperature in dark filled bottles was within the range of references values, Balbaa (1981), Soad (1973) and El-Zahwey (1992).

G.L.C. analysis was conducted to identify the compounds of these oils where citral was the main component which increased to 42.21% as shown in Fig. (6). It could be mentioned that lemongrass oils are considered available for marketing after 360 days of storage in room temperature.

Dark bottles filled with oils obtained after different times of steam distillation were stored in a refrigerator at 5°C for 360 days. The oils were investigated each 90 days and results obtained are shown in Table (4) and Fig. (7).

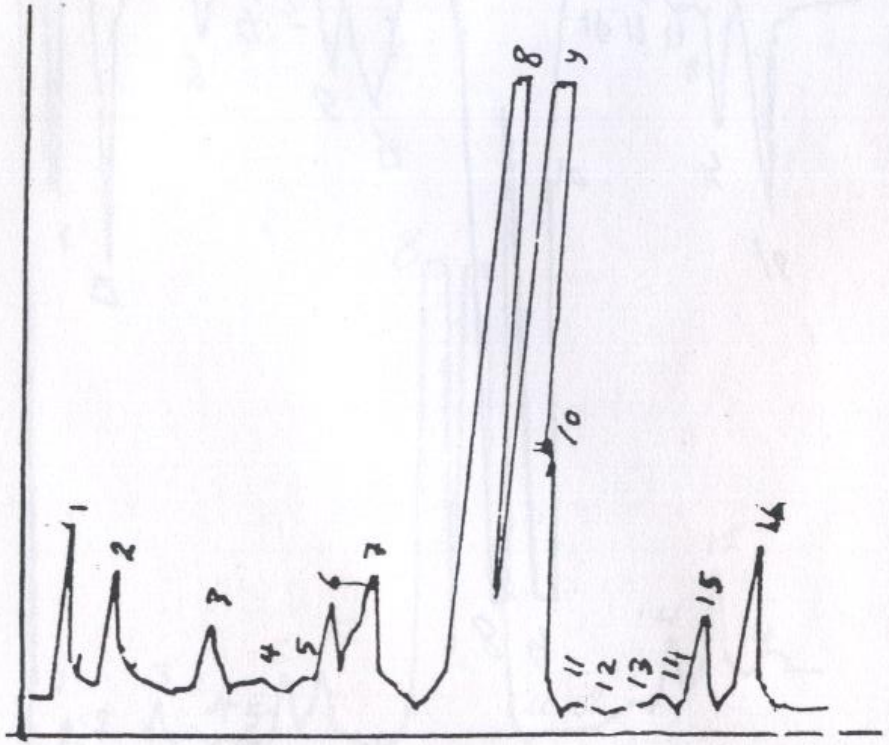


Fig. (1): Gas chromatogram of lemongrass oil sample produced after 1 hr of distillation.

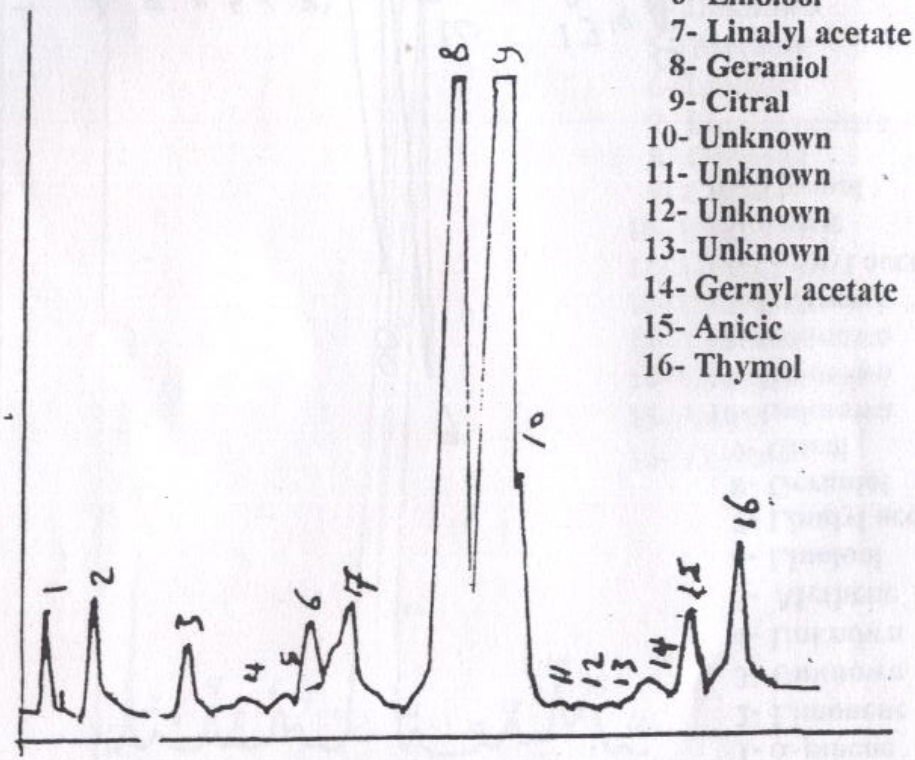


Fig. (2): Gas chromatogram of lemongrass oil sample produced after 2 hr of distillation.

Peak no.	Compounds
1	α-pinene
2	Limonene
3	Unknown
4	Unknown
5	Methene
6	Linolool
7	Linalyl acetate
8	Geraniol
9	Citral
10	Unknown
11	Unknown
12	Unknown
13	Unknown
14	Gernyl acetate
15	Anicic
16	Thymol

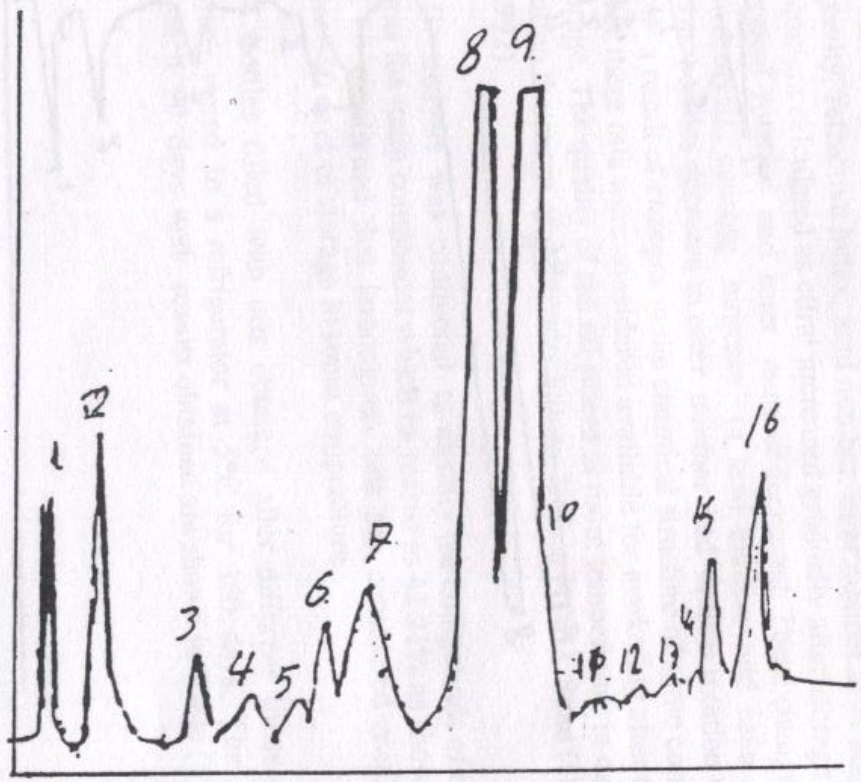


Fig. (3): Gas chromatogram of lemongrass oil sample produced after 3 hr of distillation.

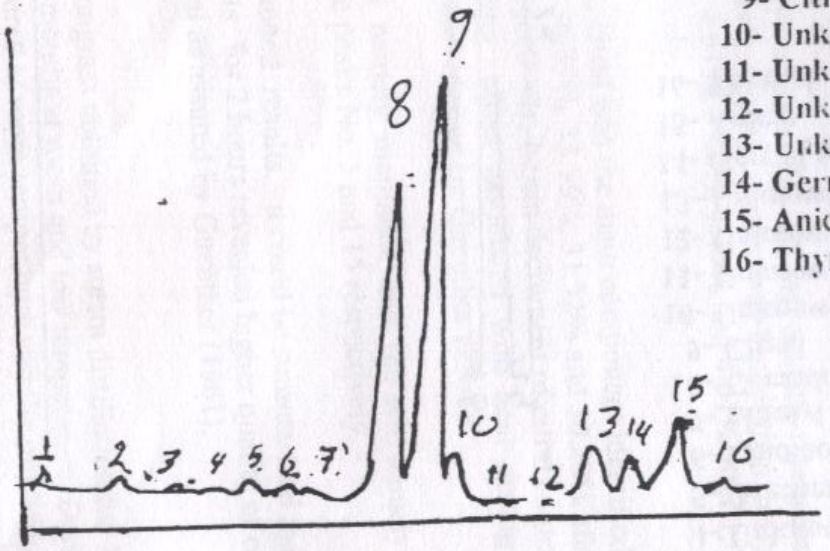


Fig. (4): Gas chromatogram of lemongrass oil sample produced after 4 hr of distillation.

Peak no. Compounds

1- α-pinene
2- Limonene
3- Unknown
4- Unknown
5- Methene
6- Linolool
7- Linalyl acetate
8- Geraniol
9- Citral
10- Unknown
11- Unknown
12- Unknown
13- Unknown
14- Gernyl acetate
15- Anicic
16- Thymol

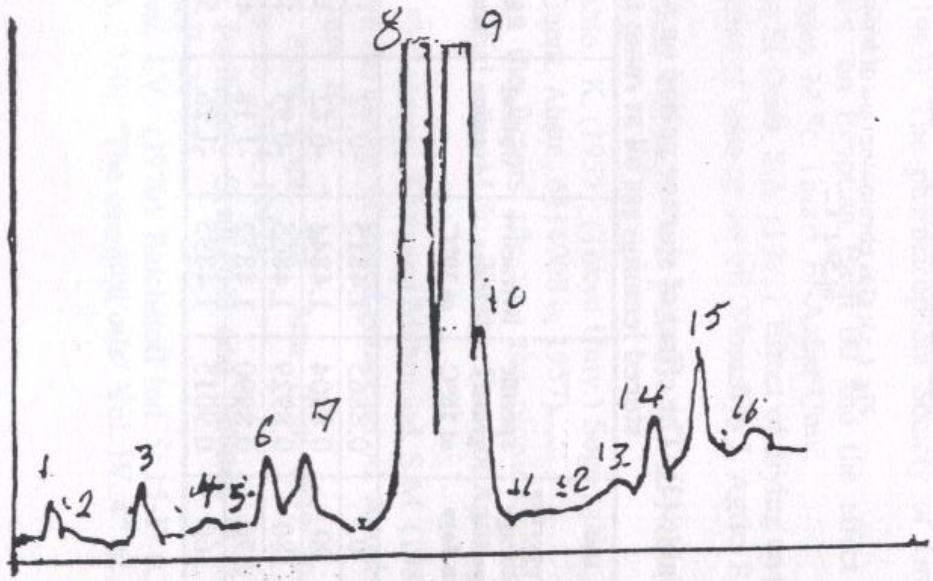
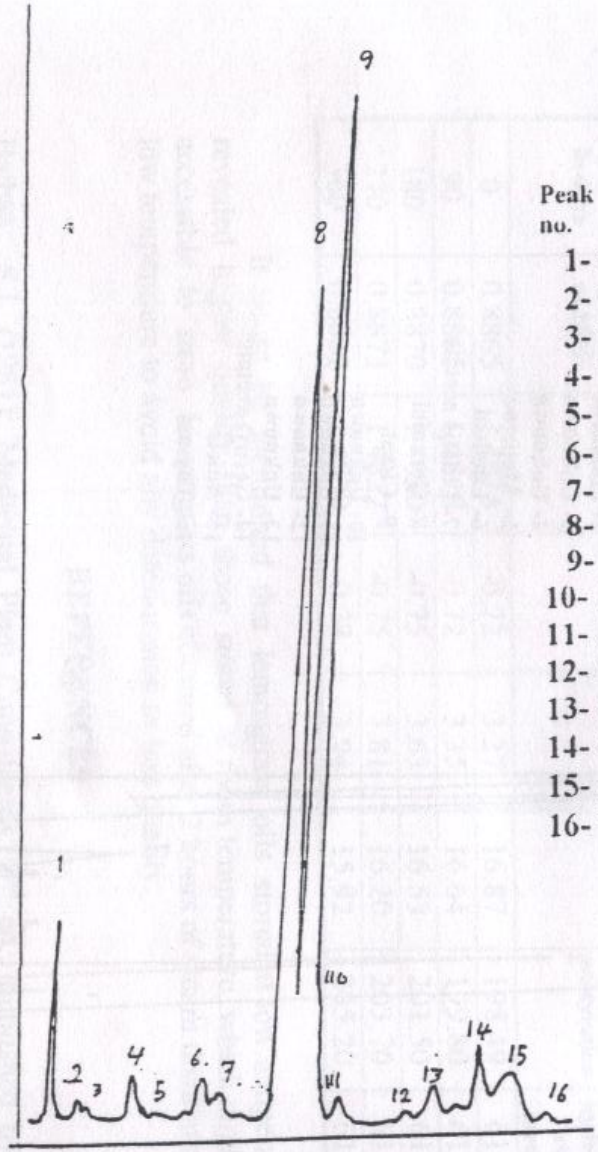


Fig. (5): Gas chromatogram of lemongrass oil sample produced after 5 hr of distillation.



Peak no. Compounds

1- α-pinene
2- Limonene
3- Unknown
4- Unknown
5- Methene
6- Linolool
7- Linalyl acetate
8- Geraniol
9- Citral
10- Unknown
11- Unknown
12- Unknown
13- Unknown
14- Gernyl acetate
15- Anicic
16- Thymol

Fig. (6): Gas chromatogram of lemongrass oil stored at room temperature.

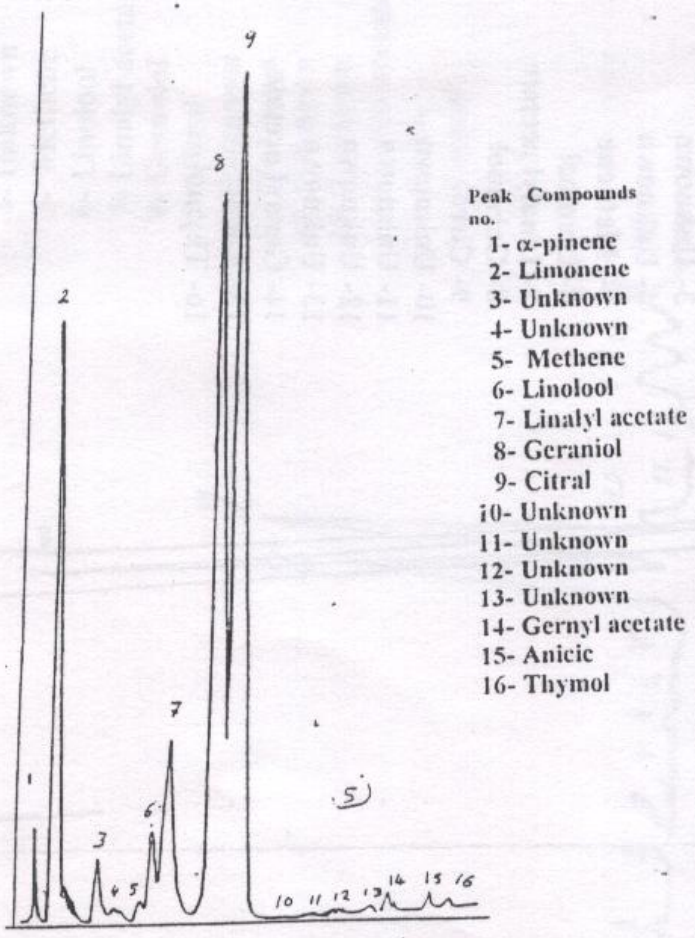


Fig. (7): Gas chromatogram of lemongrass oil stored at 5±2°C.

Table (3): The effect of storage period on physicochemical properties of stored lemongrass oil at room temperature.

Properties storage period in days	Specific gravity at 15°C	Refractive index at 20°C	Optical rotation	Acid number	Ester number	Ester No. after acetylation	Aldehyde content as citral (Hydroxyl amine hydrochloride method)
0	0.8865	1.4815	-0.72	3.27	16.87	198.49	61.44
90	0.8904	1.4844	-0.82	3.38	16.42	202.53	61.58
180	0.8929	1.4865	-0.95	4.13	15.60	211.40	61.90
270	0.8990	1.4872	-1.18	4.35	15.24	218.20	62.30
360	0.9015	1.4895	-1.26	5.22	14.85	228.49	62.82

Table (4): The effect of low temperature on physicochemical properties of stored lemongrass oil.

Properties storage period in days	Specific gravity at 15°C	Refractive index at 20°C	Optical rotation	Acid number	Ester number	Ester No. after acetylation	Aldehyde content as citral (Hydroxyl amine hydrochloride method)
0	0.8865	1.4815	-0.72	3.27	16.87	198.49	61.44
90	0.8865	1.4818	-0.72	3.35	16.64	199.80	61.46
180	0.8870	1.4820	-0.75	3.63	16.53	201.50	61.58
270	0.8873	1.4822	-0.75	3.81	16.20	203.70	61.74
360	0.8882	1.4830	-0.79	3.94	15.92	205.20	61.88

It could be mentioned that lemongrass oils stored at low temperature revealed higher quality than those stored at room temperature, which might be accessible to store lemongrass oils obtained after 2 hours of steam distillation at low temperature to avoid any deterioration or less quality.

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تأثير مدة التقطير والتخزين على الصفات الطبيعية والكيماوية لزيت حشيشة الليمون

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تم الحصول على زيت حشيشة الليمون بطريقة التقطير بالبخار بعد الفرط مباشرة وذلك بعد ١، ٢، ٣، ٤، ٥ ساعات من التقطير ووجد أن أعلى نسبة كانت بعد ساعتين (٠,٢٨%) وتم تقدير الصفات الفيزيائية والكيميائية للزيت فى كل معاملة ووجدت أن كلها مطابقة للمواصفات وتم تقدير الألدهيدات وتم حسابها كسترال بطريقة هيدروكسيل أمين هيدروكلوريد وكانت نسبة السترال ٥٨,٣٢%، ٦١,٢٤%، ٦٠,٤٤%، ٥٩,٢١%، ٥٧,٨٣% بعد ١، ٢، ٣، ٤، ٥ ساعات على الترتيب.

وتم فصل مكونات الزيت والتعرف عليها بواسطة جهاز الكروماتوجرافى الغازى ووجدت أن نسبة السترال مطابقة للمواصفات الكيميائية وعلى ذلك ينصح بتقطير عشب حشيشة الليمون لمدة ساعتين فقط نظرا لإرتفاع نسبة الزيت ونسبة السترال وجودة الصفات الطبيعية والكيميائية وكذلك لتوفير الطاقة المستخدمة فى عملية التقطير.

وتم تخزين الزيت لمدة ٣٦٠ يوما على درجة حرارة الغرفة وعلى درجة حرارة منخفضة ٥±٢°م لدراسة تأثير التخزين على الزيت.

ووجد أن تخزين زيت حشيشة الليمون فى درجة حرارة الغرفة لفترة طويلة سبب زيادة تدريجيه فى الكثافة النوعية ومعامل الإنكسار – الحموضة – رقم الإستر بعد الإستلة – الألدهيدات فى حين أن رقم الإستر والدوران الضوئى ينخفضان بإضطراد وعموما يكون الزيت صالح التسويق بعد ٣٦٠ يوميا.

وتخزين زيت حشيشة الليمون على ٥±٢°م أبطأ من معدلات التغير فى معظم الخواص الطبيعية والكيميائية مما يجعل الزيت صالح للاستخدام حتى بعد ٣٦٠ يوما وهى مدة الدراسة.