OPTIMIZATION STUDIES ON LEMON GRASS OIL EXTRACTION BY STEAM DISTILLATION

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Abstract
Due to the medicinal value of Lemongrass oil, it is widely extracted from lemongrass using a separation technique. With this method, lemongrass oil is obtained as distillate preserving the natural qualities of the plant. Distillation consisting of the steps boiling, condensing and decantation is taken up on a laboratory scale. Gas Chromatography (GC) is employed to analyse the lemon grass oil, to evaluate oil for its 'citrals' content (Gernial and Neral) qualitatively and quantitatively. The vapours emanating out of distillation are condensed to obtain a two phase system of oil and water, which is further decanted. The present paper discusses the variations in yield and specific steam consumption with respect to density of grass in the distillation flask. Also, the kinetics of extraction was studied.

Keywords: Lemongrass; Essential Oil, Steam Distillation; Citrals, Kinetics

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Introduction:

The process of extracting essential oil from plant material involves steam distillation. For heat-sensitive materials like oils, resins and, hydrocarbons, etc., it is a specific kind of distillation. These substances can decompose at higher boiling temperatures despite normally being insoluble in water. The essential property of steam distillation makes it possible to distil a chemical or a mixture of compounds at a temperature that is far lower than the boiling point of the separate components. Components of essential oil have boiling points of 200ºC or higher. However, these components volatilize in the presence of steam or boiling water at a temperature of 100ºC and 1 atmospheric pressure. Gas Chromatography is used to analyse essential oil. To determine the components in the oil and the proportion of those components, respectively along with the purity of that particular oil, qualitative and quantitative analyses are conducted.

Most tropical nations have lemongrass (*Cymbopogon citratus*) plants that thrive as herbs. It is a member of the Cymbopogon genus of aromatic grasses and has a wonderful lemon-flavoured essential oil. According to studies, citral, containing more than 75% (w/w) of essential oil (Lewinsohn et al. [1]; Luiz et al. [2]), is the main component of lemongrass. Citral is the name given to the naturally occurring mixture of two isomeric acyclic monoterpene aldehydes, geranial (trans citral, citral A) and neral (cis-citral, citral B), in a 4:1 weight ratio.

Essential oil from lemongrass revitalizes the body, clears headaches due to jetlag and relieves the body from stress and nervous exhaustion (Marongiu et al. [3]). It is known to treat respiratory infections such as sore throat, laryngitis and fever. It is also used as food additive and is a popular food flavoring agent among Asians. There are only few studies reported on the lemongrass oil extraction; most of them applied steam distillation (or hydro-distillation) process. Lewinsohn et al. [1] mentioned the use of hydro-distillation by extracting citral from fresh leaves and was later identified by GC technique.

In a work by Luiz et al. [2], lemongrass oil is extracted using dense carobn disoxide at temperatures between 23 and 50ºC and pressure in the range of 85 to 120 bar. The samples' GC-MS analyses confirmed the presence of cuticular waxers. Compared to supercritical extracts, liquid carbon dioxide extracts included more of co-extracted waxes. After the study they proposed ideal operating conditions of 120 bar and 40ºC for the extraction operation.

Using the GC flame ionization detection method, Schaneberg and Khan [4] studied on the quantification of bioactive marker compounds in essential oils containing neral, geranial, geraniol, limestone, citronellas, and beta myrcene. For the extraction of essential oils from *C. citratus,* four processes solvent extraction, steam distillation, accelerated solvent extraction, and supercritical fluid extraction were contrasted. Sonication-based solvent extraction with nonpolar solvents produced outcomes that were comparable to those of steam distillation. They have examined and contrasted a number of commercial products made from *Cymbopogen flexuosus and C. citrates.*

Another study by Masamba et al. [5] extracted for its oil from Lemongrass, collected from Domasi, Zomba by the hydro distillation method. The fresh weight oil content was 0.211±0.094%, whereas the moisture content and specific gravity were 73.28±4.98% and 0.8862±0.0071 g cm^{-3} respectively. The following compounds of lemongrass oil were identified by gas chromatography: (i) geranial (41.67%) (ii) Neral (40.33%) (iii) myrcene (9.99%) (iv) borneol (1.62%) (v) methyl-2.4-decadienoate (1.28%), and (vi) geranyl acetate (0.95%). Lemongrass oil extracted from Malawi possess chemical qualities and composition that are similar to those described in published works. On LGB (Prostephanus truncatus), the oil has fumigant and contact toxicity effects.

For the steam distillation of lemongrass, Koul et al [6] developed a first order kinetic model that was tested on pilot scale equipment using lemon grass batches ranging from 70 to 1000 kg. Their study showed that increase in the yield is observed by loosely packing the plant material inside distillation still.

Zaibunnisa et al. [7] successfully optimized the extraction of lemongrass oleoresin by Pressurized Liquid Extraction (PLE) method. Neral, geraniol, and geraniol, which together make up 72% oleoresin, were measured using the GCMSD during the study. The ideal working parameters for PLE were reported as 167ºC, 1203 psi, and a static time of 20.43 min based on maximal chemical extraction. Hydro distillation and solvent extraction were employed for comparison between PLE extract and traditional extraction techniques.

In this paper an attempt is made to optimize the parameters viz., density of the grass, and steam consumption too get the best % yield of oil (w/w). And this study was conducted both on a partially wet grass and dried grass. The paper also attempts to study the kinetics of extraction and propose a rate equation for the extraction process.

Experimental set-up:
Fig.1 depicts the steam distillation experimental setup. Experiments were carried out on different packing densities ranging from 50 to 250 gms. The lemon grass (chopped) is weighed and filled into a three necked round bottomed flask of 2000 ml capacity which is connected to a 1000 ml steam generating round bottomed flask. A double-pipe condenser is also connected to the distillation still. The condenser is further connected to a burette for separation. With the help of a burner the steam generating flask is heated and the steam generated is passed into the distillation still.

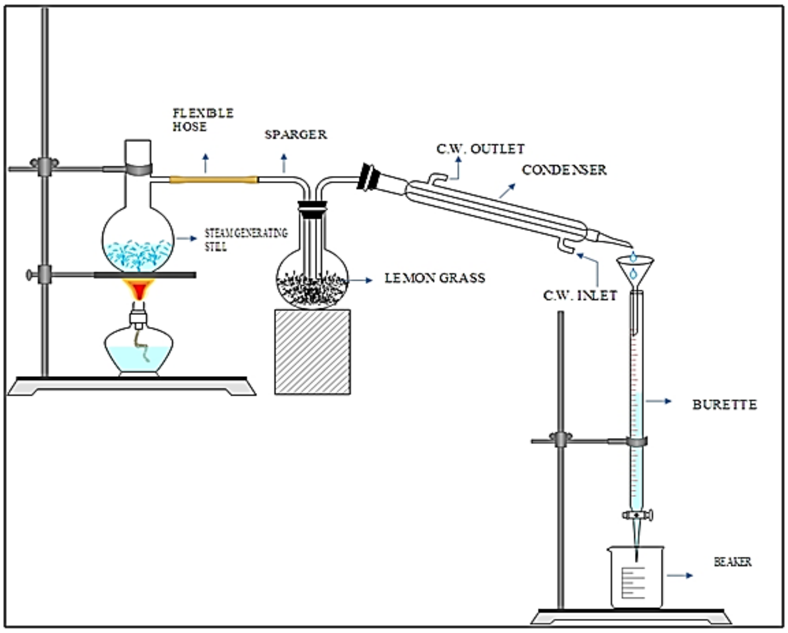

Figure 1: Laboratory Set up

The steam percolates through lemon grass, carrying the oil from grass along with it. This mixture of oil and steam passes into the condenser. The vapours condense here, and the oil and water are collected in the burette, where the water is separated. The oil produced has a light yellow colour and a sweet citratus fragrance.

Results and discussion:
Two sets of experiments were conducted, one on partially wet grass and the other with dry grass. Table 1 represent the first set of experiment, and Table 2 represent the second set. The weight of lemongrass taken in 2 lts capacity round bottom flask ranges from 50 to 250 gms. Comparing Tables 1 and 2, oil yield in the Table 1 is smaller.

Table 1. Yields of oil from lemongrass which is partially wet.

Exp No.	Weight of grass (gms)	condition	Packing	Oil Collected (ml)
1.	50	Chopped	Loose	0.8
2.	100	Chopped	Moderate	1.4
3.	150	Chopped	Moderate	2.5
4.	200	Chopped	Tight	2.7
5.	250	Chopped	Tight	3.0

Table 2. Yields of oil from lemongrass which is DRY.

Exp No.	Weight of grass (gms)	condition	Packing	Oil Collected (ml)
1.	50	Chopped	Loose	1.2
2.	100	Chopped	Moderate	2.5
3.	150	Chopped	Moderate	4.2
4.	200	chopped	Tight	3.8

Each experiment used steam distillation for a maximum of one and half hours. The oil produced was recorded after every 10 minutes in both sets of experiment for kinetic studies. Kinetic study, % yield of oil and steam consumption for oil extraction is carried out for the two sets of experiments viz., with partially wet grass and dry grass.

Experiment 1: With Partially wet grass

Table 3, Table 4 and Table 5 represent the yield of oil with respect to time, % yield of oil and specific steam consumption for different packing densities (weight of grass in grams per 2 lts volume) of lemon grass respectively. Figures 2, 3 and 4 depict the trend of the oil yield with respect to time, the oil yield percentage, and specific amount of steam consumed for various packing densities of lemon grass

Table 3. Yield of oil with respect to time for different packing densities of lemon grass
(The weight in each column indicates weight of grass packed in 2 lts round bottomed flask)

S.No	Time (minutes)	Oil(ml) 50gms	Oil(ml) 100gms	Oil(ml) 150gms	Oil(ml) 200gms	Oil(ml) 250gms
1.	0	0	0	0	0	0
2.	10	0.1	0.3	0.4	0.5	0.1
3.	20	0.5	0.7	1.2	1.3	0.4
4.	30	0.7	1.0	1.7	1.9	0.9
5.	40	0.8	1.3	2.0	2.2	1.8
6.	50	0.8	1.3	2.2	2.3	2.5
7.	60	-	1.4	2.3	2.5	2.8
8.	70	-	-	2.5	2.6	2.9
9.	80	-	-	-	2.7	3.0
10.	90	-	-	-	2.7	3.0

From Table 3, it is observed that for different packing densities of grass taken that is. for 50, 100, 150grams, the yield of oil (ml) is increased by 0.8 at each step. For further increase in packing density, there is a decrease in the yield (ml). Time taken for extraction of oil will be more for packing density above 150 grams of grass. Also it is observed that the oil extracted decreases beyond 150 grams of packing density. Since the packing density becomes tight, the steam cannot pass through the overall grass, so there will be a decrease in the yield (ml) of oil.

In Table 4, the oil yield percentage with respect to weight of grass for different packing densities is represented. Here it is observed that the yield percentage is nearly same for 50 and 150 grams of packing density. The percentage of yield decreases for 200 grams and further decreases for 250 grams. The decrease in the trend is shown in the Fig. 3.

Table 4. Wt of grass (gms) vs % Yield (w/w)

S.No	Wt. of grass(gms)	Gross yield(ml)	%Yield (w/w)
1.	50	0.8	1.42
2.	100	1.4	1.24
3.	150	2.5	1.48
4.	200	2.7	1.20
5.	250	3.0	1.07

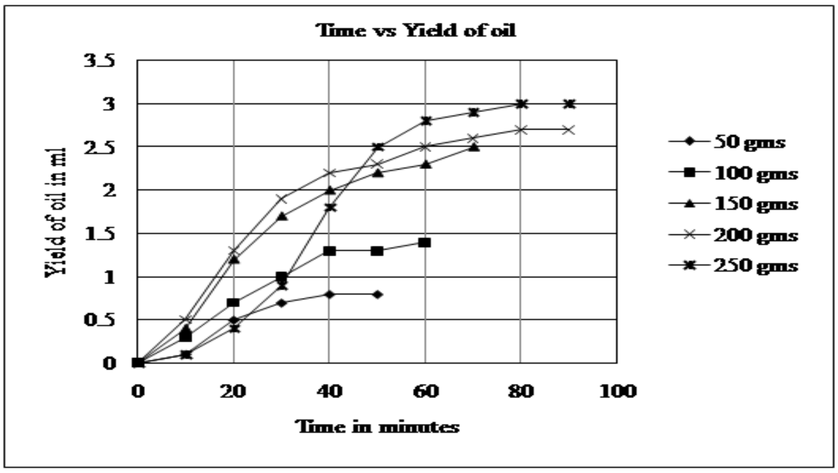

Figure 2: Time (minutes) vs Yield of Oil

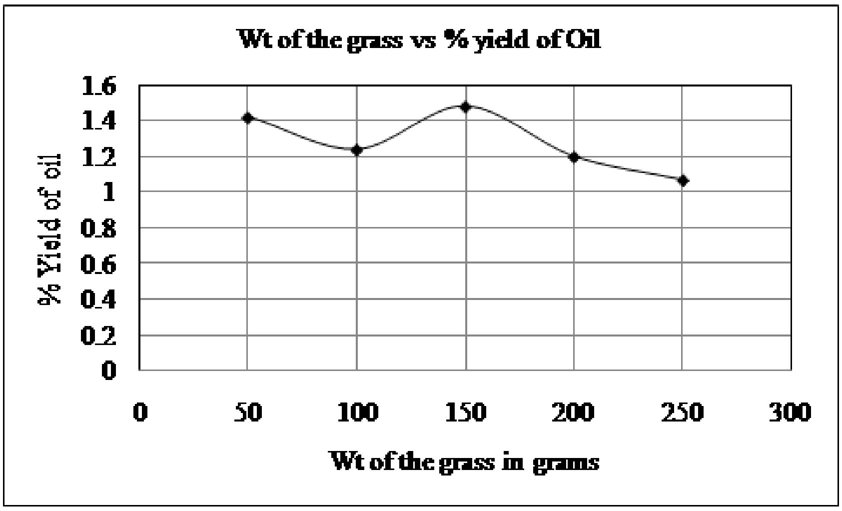

Figure 3: Wt. of the grass (gms) vs Yield %

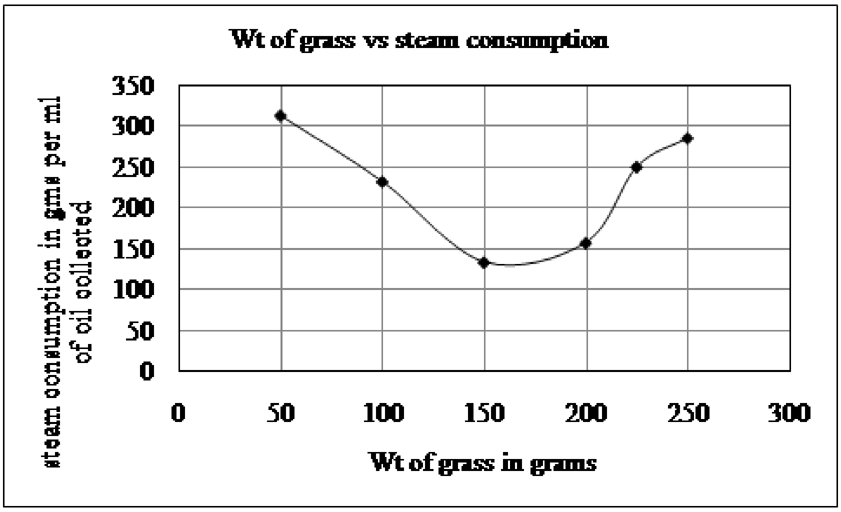

Figure 4: Wt. of grass (gms) Vs specific steam consumption per ml of oil collected

From Table-5 and Fig.4, we observe that the steam consumed is the lowest for 150 grams of grass. From this we can also say that, at 150 grams the steam consumed is low, so it is said to be optimum.

Table 5. Wt. of grass (gms) vs specific Steam Consumption per ml of oil collected

S.No	Wt. of grass(grams)	Steam consumed (per ml of oil collected)
1.	50	312.5
2.	100	232.14
3.	150	134.00
4.	200	157.40
5.	225	250.00
6.	250	285.00

Experiment 2: With Dry grass

Table 6, Table 7 and Table 8 represent the yield of oil with respect to time, % yield of oil and specific steam consumption for different packing densities of lemon grass respectively. Figures 5, 6 and 7 depict the trend of oil yield with regards to time, oil yield as a percentage, and specific steam consumption for various densities of lemon grass packing.

From the Table 6 and Fig .5, it is observed that for 150 gms of grass, maximum amount of oil is obtained. The time taken for extraction of oil is approximately same for 50, 100 and 150 grams of grass and time increases for packing density of more than 150 grams. Also, oil extracted decreases for weight of grass more than 150 grams.

Table 6. Represents yield of oil with respect to time for different packing densities of lemon grass (The weight in each column indicates weight of grass packed in 2 lts round bottomed flask)

S.No	Time (minutes)	Oil(ml) 50gms	Oil(ml) 100gms	Oil(ml) 150gms	Oil(ml) 200gms
1.	0	0	0	0	0
2.	10	0.5	1.1	2.3	1.9
3.	20	0.9	1.8	3.3	2.8
4.	30	1.0	2.1	3.7	3.4
5.	40	1.1	2.3	4.0	3.6
6.	50	1.2	2.5	4.2	3.7
7.	60	1.2	2.5	4.2	3.8
8.	70	1.2	2.5	4.2	3.8

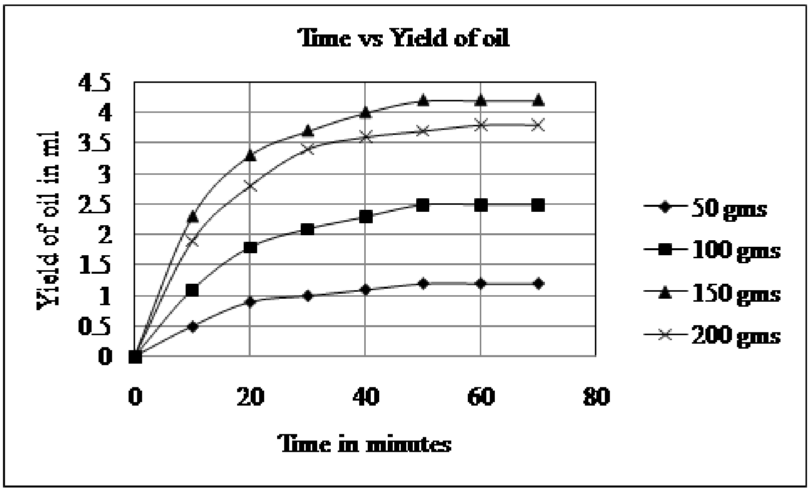

Figure 5: Time (minutes) vs Yield of Oil

Table 7 depicts that the % yield of oil is approximately equal for 50, 100, 150 grams of grass. %Yield decreases beyond the limit of 150 grams of packing density. The decreasing trend in % yield is also shown in the Fig. 6.

Table 7. Amount of yield with respect to weight of grass

S.No	Wt. of grass (grams)	Gross Yield(ml)	%Yield (w/w)
1.	50	1.2	2.136
2.	100	2.5	2.225
3.	150	4.2	2.492
4.	200	3.8	1.691

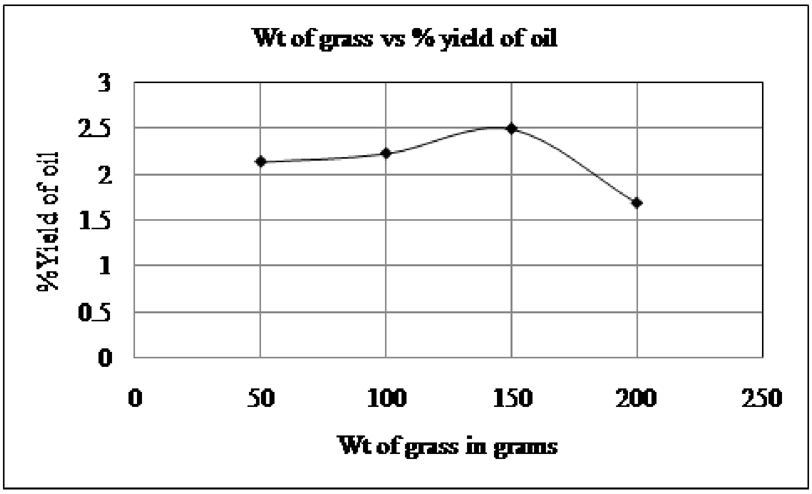

Figure 6: Wt. of the grass (gms) vs Yield %

Table-8 and Fig 7 depicts that the specific steam consumed is the lowest for 150 grams of grass. As the steam consumed is low with 150 gms of lemongrass, so it is said to be optimum.

Table 8. Represents steam consumption with respect to weight of grass

S.No	Wt. of grass (grams)	Steam consumption (per ml of oil collected)
1.	50	133
2.	100	96
3.	150	83
4.	200	143.15

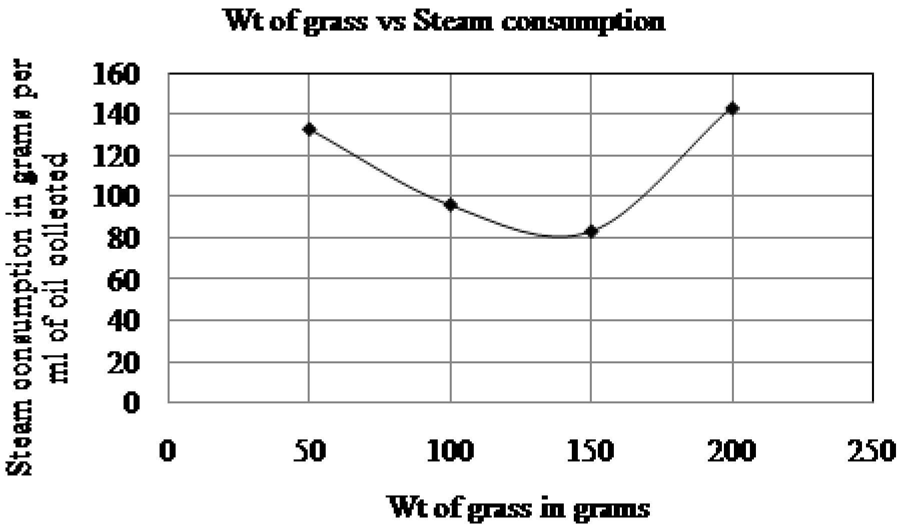

Figure 7: Wt. of grass (gms) vs specific steam consumption per ml of oil collected

Theory and Kinetic Studies:
Steam's direct contact heat transfer first makes grass wettable at high temperatures, promoting the osmosis and diffusion of the oil. Lemon grass essential oils extracted by steam distillation is a rate process. Oil removed per unit time is directly proportional to oil left over in the grass. First order kinetics provides the simplest rate equation (Koul et al. (6)),

$$-\frac{dm}{dt} = k \quad (1)$$

$$-\frac{dm}{m} = kdt \quad (2)$$

By integrating Eq (2) we obtain,

$$\ln(\frac{m_0}{m}) = k.t \quad (3)$$

$$\ln(\frac{1}{1-u(t)}) = k.t \quad (4)$$

where
m = average essential oil content of grass at any given time t,
m_0 = initial concentration of essential oil at the beginning of steam distillation
t = time of steam distillation
u(t) = fraction of oil extracted

$$u = \frac{Oil\ extracted\ till\ time, t}{Cumulative\ amount\ of\ oil\ extracted} \quad (5)$$

If the aforementioned model is accurate, a straight line passing through the origin should result from the plot of ln (1/(1-u)) vs time. As previously noted, the tests that produced fractional yield of oil, u, are shown in the table below (Table 9).

Table 9: Kinetics of the experiment conducted

S.No	Time interval (min)	Oil collected (ml)	u	ln(1/(1-u))
1.	0	0	0	0
2.	10	2.3	0.380	0.8
3.	20	3.3	0.666	1.6
4.	30	3.7	0.88	2.12
5.	40	4.0	0.952	3.036
6.	50	4.2	1	-
7.	60	4.2	-	-

For the experiment with 150 grams of grass, ln (1/(1-u)) vs t, was plotted to confirm the rate model as stated in the theory. All the experiments with respect to 50, 100, 150, 200 grams of grass represent straight line behavior. In this paper, only the kinetics of 150 grams is represented. Fig 8 indicates a straight line behavior, which means that it is a first order kinetics. The rate constant is $K=0.08 min^{-1}$.

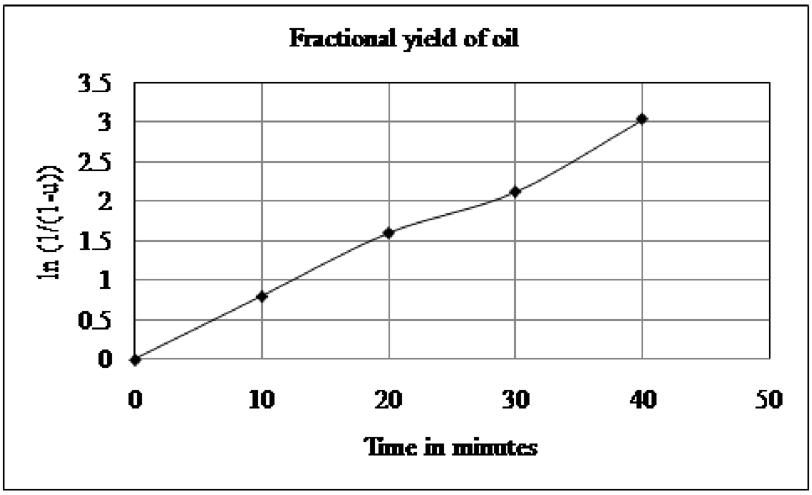

Figure 8: Time vs ln(1/(1-u))

Conclusions:

1. Loose and moderate packing that is, for packing density of 50 and 75 gm/l gives higher yield when compared to tight packing that is., for packing density of 100 and 125 gm/l.
2. Dry grass gives higher yields when compared to semi wet grass. The yield % for dry grass at 75 gm/l is 2.492 compared to yield % for wet grass at 75 gm/l is 1.483 and is higher by 65%.
3. Yield on laboratory scale is at least 80% higher than the commercial extraction units in the vicinity of Visakhapatnam.
4. Yield % is optimum for packing density of 75 grams per 1 litre volume and yield decreases beyond this limit.
5. Steam consumption and time of extraction is also lowest for 75 gm/l packing density.
6. First order kinetics are shown by the plot of ln (1/ (1-u)) vs t, which indicates straight line behavior.
7. A modification in the present design of commercial steam distillation unit can be taken up based on the experimental results obtained in this paper, in order to improve their overall performance.

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