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Modelling the Kinetics of Steam Distillation of Essential Oils from Lemon Grass (Cymbopogon Spp.)

Amenaghawon Nosakhare Andrew*, Okhueleigbe Kenneth Ehimare,
Ogbeide Samuel E., and Okieimen Charity O.

Department of Chemical Engineering Faculty of Engineering University of Benin, Benin City, Nigeria

Abstract: The kinetics of essential oil recovery from lemon grass was investigated using an experimentally validated first order kinetic model. Essential oil recovery was accomplished in a steam distillation equipment using 100g of plant material in different physical states. The kinetic model developed did not take into consideration the resistance to oil diffusion from the tissues within the plant material to the leaf surface. The integral method of analysis was used to test the fit of the model to the experimental results. The straight lines obtained indicated that the recovery of essential oils from lemon grass followed first order rate kinetics with a mean rate constant, *k*, of 0.0451. It was observed that oil extraction was not instantaneous as some time was required to wet the plant material to enable the oil diffuse from within the leaves to the surface. Furthermore, loose packing of the plant material within the steam distillation equipment improved oil yield and drying the plant material prior to distillation did not reduce the quantity of oil available for recovery.

Keywords: Essential oil; steam distillation; lemongrass; kinetic model.

1. Introduction

Essential oils are made up of highly volatile substances which can be extracted from numerous plant species. The oil usually bears the name of the plant from which it is extracted and a host of methods exist for separating these oils from the various plant materials [1]. The method of extraction is important in that the composition of essential oils is somewhat dependent on the extraction method employed [2]. Distillation based recovery processes such as steam and vacuum distillation are preferred for the extraction of essential oils from plant materials. This is because these processes are flexible, versatile, do not lead to the decomposition of the essential oils and provide the possibility of operating with small volumes [3].

Essential oils have an unexpected large range of applications finding uses in the food and beverage industry, the perfume industry, in the production of flavours and spices, in the soap and detergents industry, in the pharmaceutical industry, and in the plastics industry to mention a few [4, 5].

Virtually all naturally occurring essential oils are made up of hydrocarbons such as camphene, pinene, limonene, phellandrene, cedrene, and oxygenated hydrocarbons such as alcohols, aldehydes, esters, ethers, ketones, lactones, phenols and organic acids [6, 7]. The essential oils get their odours from the oxygenated compounds contained in them and these confer stability on

* Corresponding author; e-mail: andrew.amenaghawon@uniben.edu
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the oil against oxidation. The hydrocarbons are less stable and they are responsible the degradation observed in the oils [6]. The hydrocarbons contained in the essential oil are often removed so that the oils can be more stable and still retain their odours and flavours.

Lemon grass oil is a clear liquid with a strong lemon-like odour and a pale yellow colour. It is the most common and cheapest essential oil commercially available. It is obtained from steam distillation of lemon grass (*Cymbopogon* spp.). The total amount of essential oils present in lemon grass varies between 0.28-1.4% with the maximum recovered recorded as 3% [8].

In many industrial applications, mathematical models of distillation systems are frequently used to design effective control systems, train operating personnel and handle fault diagnostics [9]. Benyoussef et al. [10] reported the modelling of steam distillation of essential oils from coriander by using two diffusional models that considered both diffusion and transfer of species. Cassel and Vargas [11] simulated steam distillation of lemon grass using a model based on Fick's law in steady state for one-dimensional rectangular geometry. Ha et al. [12] developed a two-factor interaction model for the extraction of essential oil from lemon grass and a linear model in terms of coded variables for the extraction of essential oil from lemon grass stems.

In this work, we develop a simple kinetic model for the extraction of essential oils from lemon grass. A first order kinetic model equation was proposed and was subsequently validated using experimental data.

2. Materials and methods

Fresh lemongrass was procured from a local supplier in Ugbowo, Benin City, Edo State, Nigeria. The stem was cut 10 cm from root and further cut to 2 mm diameter prior to drying. The initial moisture content of the lemongrass was 11%. The samples to be used for experiments were oven dried at 40°C overnight and kept in a sealed bag. One hundred grams (100g) of lemon grass (chopped or unchopped) was charged into steam distillation equipment. The equipment was fitted with a tight lid to prevent oil and vapour from leaking out. The equipment functioned in such a way that steam rising from the still stripped the oil away from the plant material and the vapour comprising oil and steam was passed to a condenser where it was condensed and separated. The oil was separated from the water at the top of the separator as it is less dense than water which was removed from the bottom of the separator.

3. Mathematical modeling

The extraction of essential oils from lemon grass by steam distillation has been reported to be a kinetic process [6]. This means that the rate at which oil is removed from the plant material is entirely dependent on the average quantity of essential oil present in the grass at any time. For the modeling exercise done in this work, the diffusion of the oil from the tissues within the leaves to their surface was not considered. The modeling effort is only focused on the transport of oil from the surface of the leaves into the vapor phase. The rate at which oil is extracted from lemongrass is directly proportional to the amount of oil present in the plant material at any time *t*. This was expressed mathematically in the form of a first order ordinary differential equation (ODE).

$$\frac{dC}{dt} = -kC \tag{1}$$

where

C = average quantity of essential oil in the grass at any time t
k = first order rate constant
t = time of distillation

This ordinary differential equation was solved using the variable separable method of solving first order ODEs [13].

By separating variables, we arrive at:

$$\frac{dC}{C} = -kdt \tag{2}$$

Both sides of Equation (2) were integrated within limits C_o to C for the left-hand-side and 0 to t for the right-hand-side to obtain:

$$\ln\frac{C}{C_o} = -kt \tag{3}$$

where
C_o = initial quantity of essential oil.

The initial quantity was approximated to be a function of the cumulative volume of oil extracted from the lemon grass.

A new variable y(t) representing the time dependent fractional extraction yield was defined as follows.

$$y(t) = \frac{C_o - C}{C_o} \tag{4}$$

Equation (3) was rewritten in favour of y(t) to obtain:

$$\ln\left[\frac{1}{1-y(t)}\right] = kt \tag{5}$$

Equation (5) is the solution of the ODE presented in Equation (1) and this was fitted to the results obtained from experiments to determine its validity.

4. Results and discussion

Figure 1 to 4 show the time trajectories of essential oil recovery from lemon grass. These Figures show the cumulative amount of oil in milliliters, extracted as a function of time. The results are for one hundred grams (100 g) of grass in the various states investigated. The trend observed indicates that the condition of the material prior to steam distillation did not affect the trajectory as the same trend was observed for all four curves. Each curve describes an initial almost linear increase in the cumulative volume of oil with time, the curve then flattens out about a constant value. This constant value has its mean, estimated from all results, as approximately 0.33 mL (Table 1). This amount is not an adequate representation of the total volume of oil that is actually present within 100g of lemon grass that was used in the experiments. Table 1 gives a summary of the maximum oil volumes recovered from the grass in the various states.

According to Koul et al. [6], the rate of oil vapourisation from the plant material is not affected by the relative volatility of the chemical components that make up the oil; rather it is influenced by their solubility in the vapour phase which in this case is steam. Therefore it is reasonable to conclude that some of the oil could still be trapped within the tissues of the plant inaccessible to the vapor phase. The mean value obtained is however the typical amount of oil that can be extracted using steam distillation method per 100 g of the grass.

The effect of raw material preparation as emphasized by Sterrett [14] appears not be evident in these experiments since from Figure 1 and 2, (freshly harvested chopped grass) the maximum recovery values recorded are lower than those presented in Figure 3 and 4 (freshly harvested un-chopped grass). However this trend does not disagree with that of Sterrett [14] but further strengthens it. The reduction in the size of the plant material through chopping is in fact supposed to improve oil extraction but this does not happen because of the tight packing caused by the reduction in size. Koul et al. [6] reported that tight packing of the plant material results in reduced oil extraction while loose packing improves extraction by offering less resistance to upward flow of the oil-steam mixture. Furthermore, it is possible that some of the oil might have escaped in the process of chopping the plant material. Therefore it can be inferred that between raw material preparation and packing within the still, the latter is considered as the over-riding factor. For the case of the dried grass, the result presented in Figure 4, can be explained by the fact that though the plant material is chopped and there is the occasion of tight packing within the vessel, the steam-oil mixture is still able to rise up with little or no resistance because the weight of the chopped plant material has been reduced, consequently the steam-oil mixture easily displaces the plant material as it rises up the vessel. The case presents the advantage of the combined effects of loose packing and plant preparation. The relatively high maximum volumes of oil observed for Figure 3 and 4 for dried plant material suggests that drying the plant material does not reduce the quantity of oil originally available in the plant material for extraction.

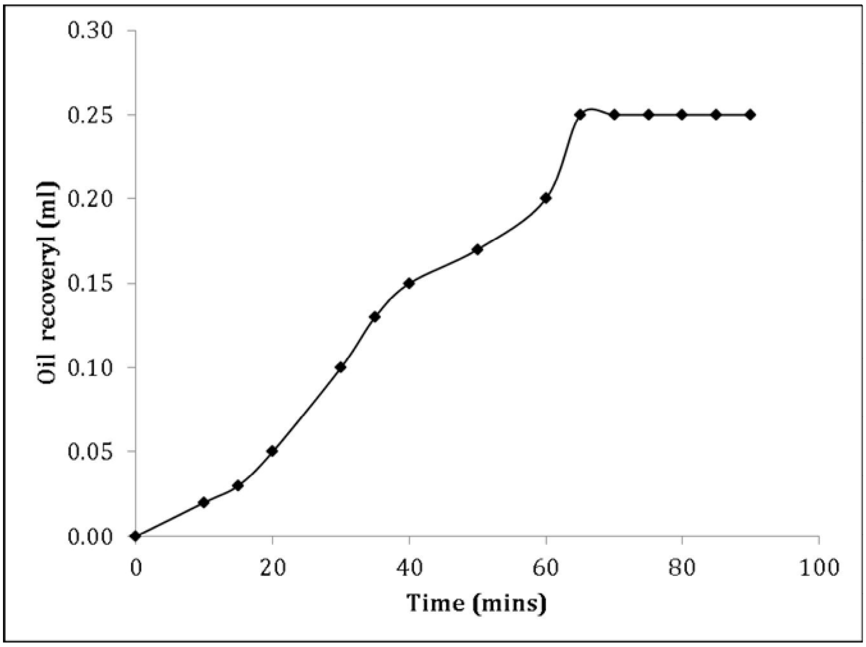

Figure 1. Exp 1: cumulative volume of oil extracted from freshly harvested chopped grass

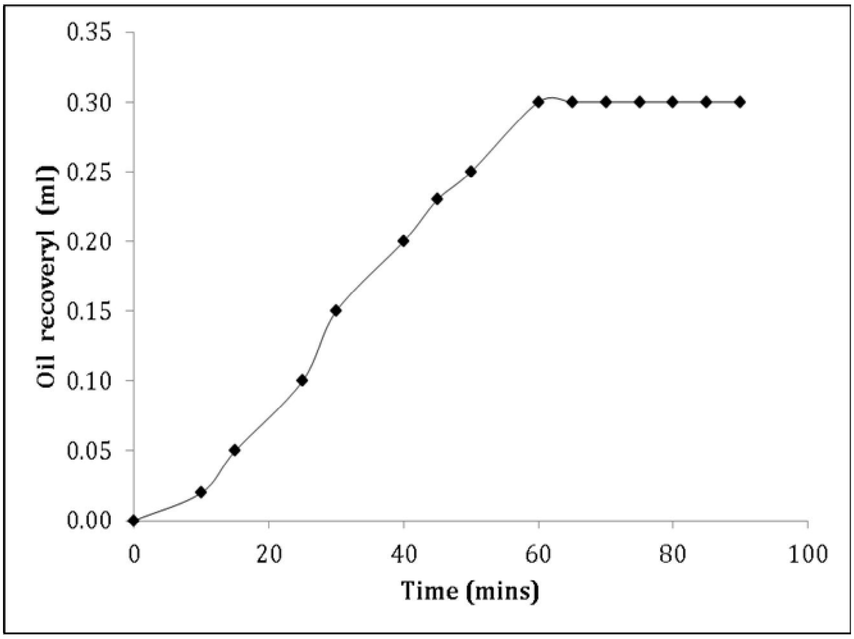

Figure 2. Exp 2: cumulative volume of oil extracted from harvested chopped grass

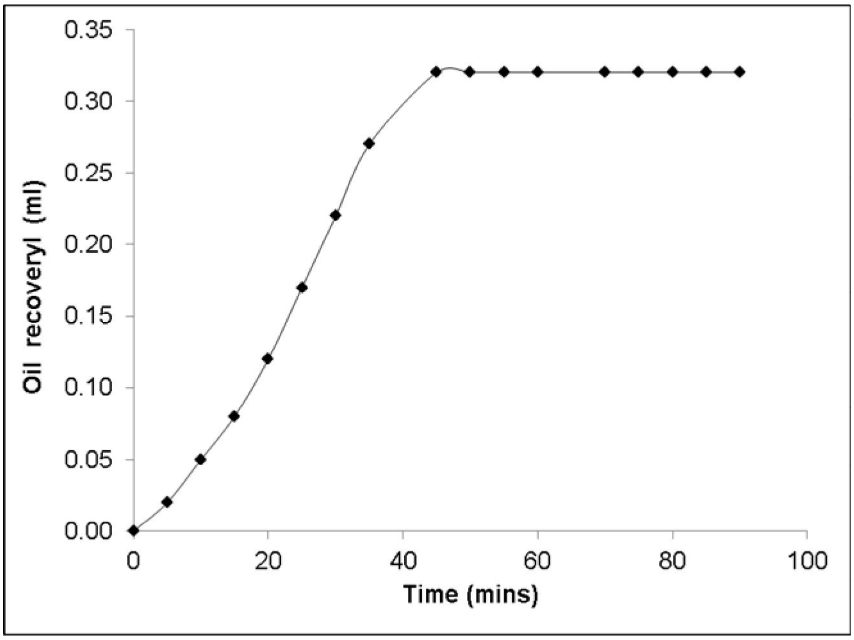

Figure 3. Exp 3: cumulative volume of oil extracted from freshly harvested un-chopped grass dried to half its weight

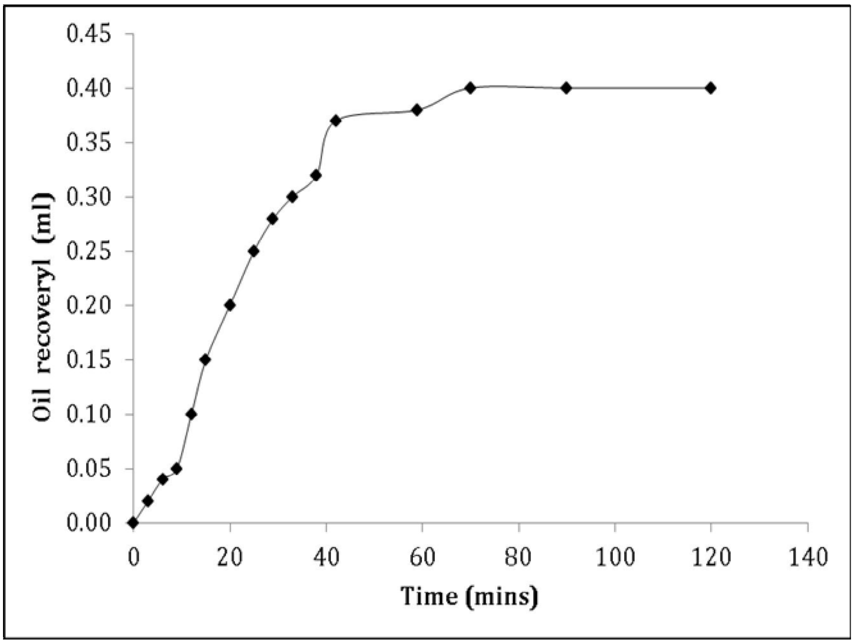

Figure 4. Exp 4: cumulative volume of oil extracted from chopped grass dried to half its weight

Table 1. Maximum cumulative oil volumes and time profile for varying raw material states

Raw material state	Maximum cumulative volume of oil (mL)
Chopped dried grass	0.40
Fresh un-chopped grass	0.32
Fresh chopped grass	0.28
Mean value	0.33

The criterion of validity of equation (5) was tested by fitting it to the experimental data. Figure 5 to 8 show the result of kinetic analysis carried out for all four experiments. For the first order kinetics proposed, a straight line should be obtained for a plot of ln[1/1-y(t)] versus *t*. It is evident from Figure 5 to 8 that the experimental results showed a good fit with the first order rate expression proposed as seen in the straight lines resulting from the plot of ln [1/1-y(t)] versus *t*. The goodness of fit is demonstrated with an average coefficient of determination (R^2) of 0.99.

Normally, the straight lines obtained should pass through the origin for first order kinetics, but that was not the case for the results presented in Figure 5 to 8 as all the lines had an intercept on the time axis. This is explained by the fact that vapourisation of the oil does not start immediately steam is injected into the still. The intercept on the time axis is actually the time it takes for the plant material to be wetted by steam before oil extraction begins. Wetting actually helps the oil to diffuse from within the leaves to the surface and once it is on the surface, it is available for stripping by steam. The kinetics is observed to follow first order after this initial wetting period. A wetting period of about 10 minutes was recorded for experiments 1 and 2 while a value of about 7 minutes was recorded for experiments 3 and 4. These results are in agreement with those obtained by Koul et al. [6]; though they recorded a wetting period of 10 to 15 minutes for 100kg of lemon grass. The slope of each of the straight lines in the plots above corresponds to the rate constant *k*, of the first order rate expression representing the vapourisation of lemon grass oil. The value of k was estimated to be between 0.0315 and 0.0566 with a mean value of 0.0451. Therefore, the rate expression for the vaporization of oil from of lemon grass can be written as:

$$\frac{dC}{dt} = -0.0451C \tag{6}$$

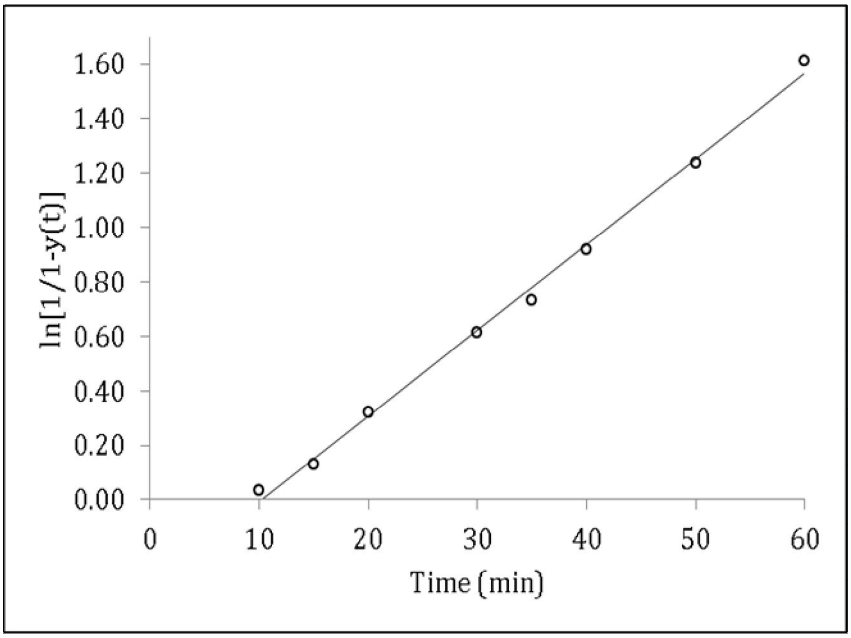

Figure 5. Exp 1: kinetic analysis for freshly harvested chopped grass

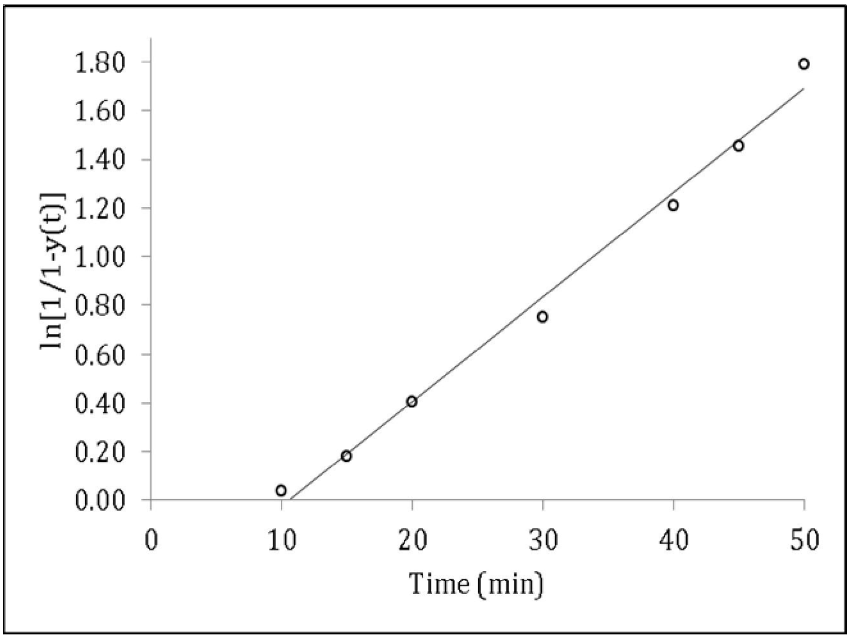

Figure 6. Exp 2: kinetic analysis for chopped grass

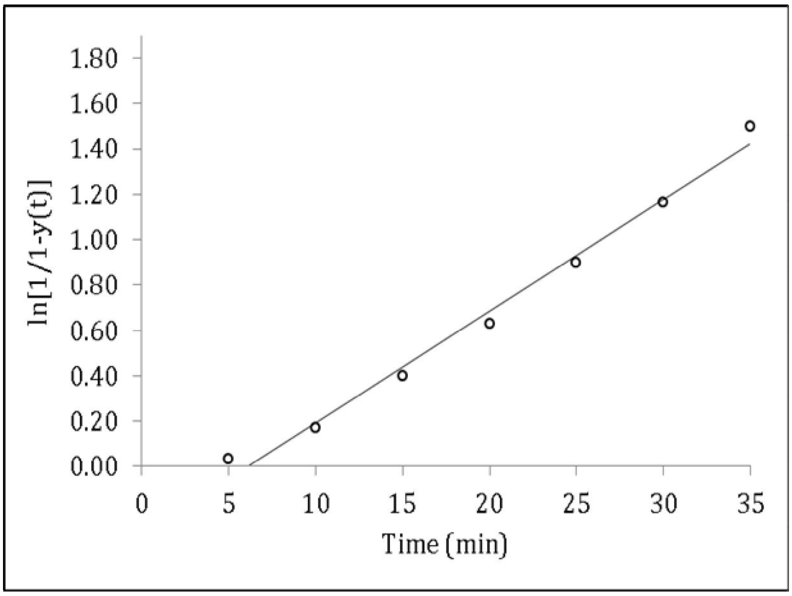

Figure 7. Exp 3: kinetic analysis for freshly harvested un-chopped grass dried to half its weight

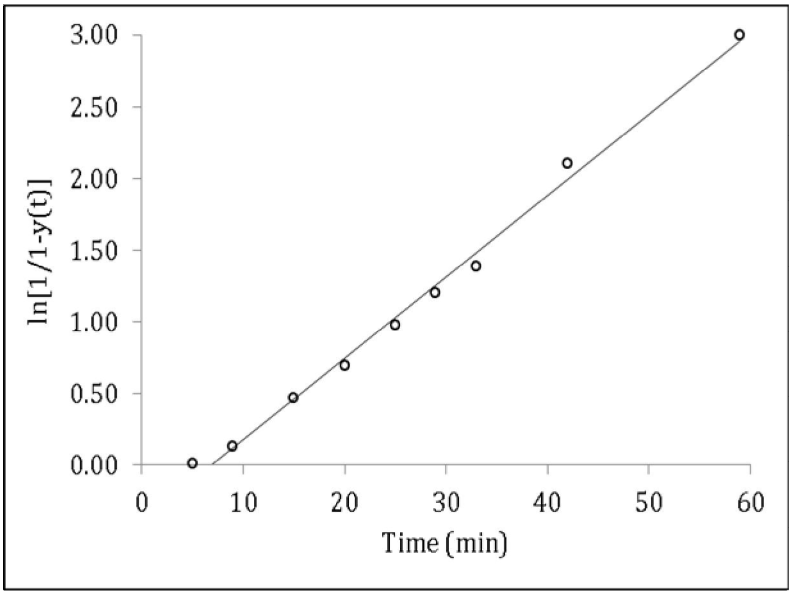

Figure 8. Exp 4: kinetic analysis for freshly harvested chopped grass dried to half its weight

5. Conclusions

In this work, the kinetics of essential oil extraction from lemon grass was investigated. The following conclusions can be drawn from this study:

a) The loose packing of plant material in the still improves the yield of oil recovery from the plant material.
b) Drying the plant material does not reduce the quantity of oil available for extraction.
c) The rate of oil vaporization from lemon grass is not influenced by relative volatility of the oil components but their solubility in the vapour phase (steam).
d) Recovery of essential oil from lemon grass by steam distillation follows first order rate kinetics with an average rate constant *k* of 0.0451. This indicates that oil extracted per unit

time is directly proportional to the amount of oil present in the grass.

e) Oil extraction from lemon grass is not instantaneous. Wetting of the plant material inside the still takes place for about 7 to 10 minutes to allow the oil to diffuse from the plant material to the surface.

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