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# Cleaner production of essential oils by steam distillation

Phineas Masango

*80 Mill Lane, Millfield Gardens, Kidderminster DY11 6YH, UK*

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**Abstract**

A new process design and operation for steam distillation of essential oils that increases oil yield and reduces loss of polar compounds in wastewater was developed. A packed bed of the raw materials, as opposed to hydrodistillation, was used. The packed bed sits above the steam source and only steam passes through it without the boiling water mixing with vegetable raw material, as is the case in hydrodistillation. The method also addresses one of the most difficult problems in steam distillation, that of determining the optimum amount of steam required for distillation of a given mass of vegetable material. The process design requires that minimum steam be passed through the packed bed so that there is minimum water in the distillate. The less the water (as aqueous fraction of the distillate) there is in the distillate, the less the water-soluble compounds that will be dissolved into the aqueous fraction of the condensate. This minimum steam flow was and can experimentally be determined. In theory, it is the steam flow rate that will develop a pressure drop with lowest steam transport across the packed bed.


*Keywords:* Extraction of essential oils; Steam distillation of essential oils; Equivalent; Linalool; Camphor; Gas chromatography of essential oils

## 1. Introduction

Although essential oils are produced by different methods such as solvent extraction, expression and critical fluid extraction, most are produced by steam distillation [1]. The proportion of different essential oils extracted by steam distillation is 93% and the remaining 7% is extracted by the other methods [7]. Essential oils are multi-component chemicals. The mixture of oil compounds that constitute the essential oil comprises polar and non-polar compounds. Some of the compounds in the composite oil are lost in the wastewaters, as shown in the work by Fleisher and Fliesher [2] and Bohra et al. [3]. In the case where the vegetable material and water are mixed in the still, the oil is lost in the water in the still as well as in the aqueous phase of the distillate. During steam distillation of essential oils, the recovery of all organic constituents as the product depends on their partition between water and oil phases of the distillate. In the majority of cases the oil is less dense than the water and so forms the top layer of the distillate. The very important compounds that make up the chemicals usually referred to, as the top notes, in the fragrance industry are the polar compounds [4,5]. This polarity makes the compounds soluble in water and this solubility is a function of the physical properties of the system such as pressure, temperature and chemical potential. In many steam distillation processes, vegetable material is mixed with water and the system is brought to a boil, a process commonly referred to as hydrodistillation [6]. The vapour is collected and condensed in order to separate the water from the oil fraction. However, the residual oil dissolved in the water usually causes odour nuisance and is also a waste of the valuable product in the water stream. Studies have been done to quantify and qualify these water-soluble compounds in distillation wastewater [6,7].

In order to optimise the recovery of essential oils, the loss of some of the oil components such as the polar components in both the aqueous fraction of the distillate and the water in the still, the water has to be redistilled, a process called cohobation. Redistilling to process wastewater in order to recover the dissolved oil components results in increased utility cost, mainly heating or energy costs [8–10].

*E-mail address:* mutape2002@yahoo.co.uk.

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## 2. Experimentation—new process design

A batch of 750 g each of artemisia leaves and lavender flowers was packed in 0.002 $m^3$ extractor at a time. The raw material forms the packed bed. The packed bed can theoretically be broken into small elements of volume from bottom to top. As steam is passed through the packed bed, it condenses in the first element before going to the next and so on, where it releases its enthalpy of vaporisation and the heat raises the temperature of solids to operation temperature. The solid vegetable particles absorb the condensate that forms when steam loses its enthalpy of vaporisation. Subsequent heat at maximum temperature is used for vaporisation/distillation of oil. In this system, presented in Figs. 1 and 2, because there are predominantly solid raw materials and steam, it is possible to analyse the mass transfer using the two-film resistance theory. In the packed bed, the vegetable biomass is not immersed in water as is the case in some steam distillation processes such as the ones reported by Reverchon and Senatore [1], Kishore et al. [4], and Perineau et al. [6].

### 2.1. Analytical method

In order to simplify the analysis of the distillation process, two essential oil sources were used for oil production using the new process design. The two sources used were *Lavendula angustifolia* (lavender) and *Artemisia annua* (artemisia). The next step was to identify one of the major components in each type of oil. Camphor was identified as one of the major components in artemisia and linalool was one of the major components identified in lavender. The components

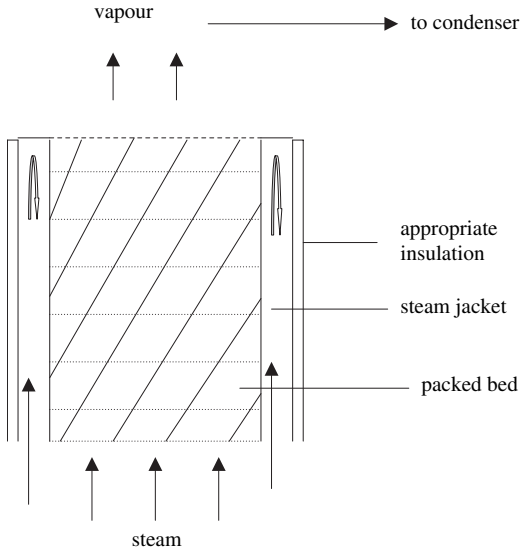


Fig. 1. Diagram of the new packed bed extraction system developed and tested to improve extraction efficiency and product quality.

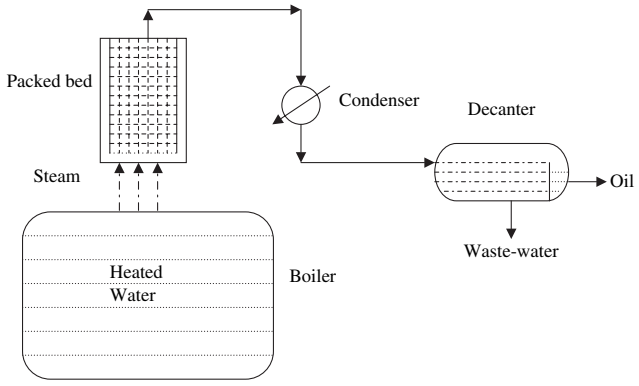


Fig. 2. Diagram of the steam distillation extraction process.

were identified using gas chromatography (GC) and the internal standardisation method. The standards were bought from Sigma Aldrich Chemicals.

### 2.2. Preparation of calibration curves

The four camphor standard solutions were prepared by dilution series. Initially 0.04 g/ml was prepared by dissolving camphor in hexane and made up to 10 ml in a volumetric flask. A sample was removed and labelled sample 1. Then 0.5 ml was removed and topped to 1 ml, which was then divided into two equal halves with one used as a dilution and the other as vial number 2. The method was repeated for the other two vials, thus making four samples. The concentrations prepared for camphor were 0.04 g/ml, 0.02 g/ml, 0.01 g/ml and 0.005 g/ml. The samples were used to draw the calibration curve shown in Fig. 3.

A similar method was used for the preparation of linalool standards. Linalool was the principal component used to develop the calibration curve for lavender oil. In this case the four vials were of concentrations 0.16 g/ml, 0.12 g/ml, 0.08 g/ml and 0.04 g/ml. The advantage of the gas chromatographic method is that these concentrations can be converted into dimensionless area and vice versa, thus, they are effective for use as calibration curves for quantifying water-soluble compounds in the distillate fractions of the different oil types such as those given in Figs. 9 and 10 using Eq. (1).

The analytical equipment used was a Perkin Elmer Autosystem GC using the FID detector. The column was BP20 type. A single temperature ramp programme was used. The programme was: injector temperature of 200 °C, oven temperature of 75 °C (2 min) oven temperature of 200 °C (2 min), detector temperature of 240 °C and the oven rate was 10 °C/min.

The two compounds used as standards in each case are of commercial interest. Camphor is used in pharmaceutical preparations and personal/skin care products and linalool is used in the fragrance industry. Both chemicals are oxygenated making them polar and

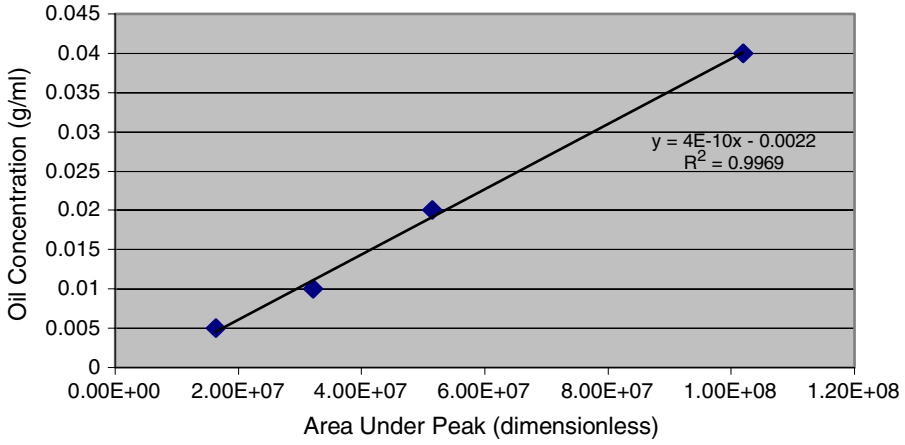


Fig. 3. Artemisia oil calibration curve using camphor-L as standard.

are the major components in water solutions of each distillate. The chromatograms of the two standard compounds were used to calculate oil yield and the amount of oil in grams of the oil dissolved and lost in the wastewater. The calibration curves were drawn using the prepared standards and temperature programme on the GC as outlined above.

## 3. Results

The calibration graphs for the two major components, one for each of the two types of oils are given in Figs. 3 and 4.

The chromatograms shown in Figs. 9 and 10 are those of the water-soluble compounds in artemisia and lavender oil, respectively. In order to determine the dissolved polar compounds in the aqueous phase of the distillate, their maximum possible content in water can be determined from the calibration curves. In order to simplify the calculations not all the minor compounds in water solution were qualitatively calculated. The composition of the minor components in water is known by their dimensionless area contribution in the respective oil chromatogram. Only the principal components were qualitatively identified and the total mass of the oil in water was corrected to take into account the other minor chemical compounds in the water solution. The calculations for the total oil, which averaged 0.24% and 0.26% (mass/volume) for artemisia and lavender oil, respectively, were based on calculations using the principal component in each case. These components are camphor and linalool in artemisia oil and lavender oil, respectively. The calculation is:

$$\text{Total oil (g)} = \frac{(\text{Total area under chromatogram})(\text{concentration as on calibration curve})}{(\text{area of principal component})} \quad (1)$$

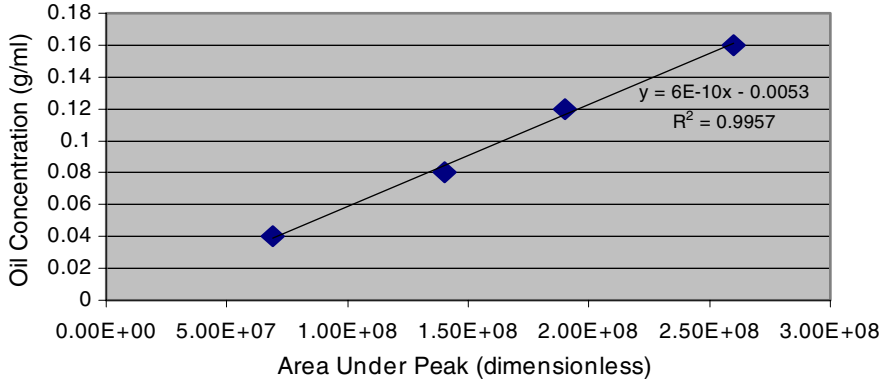


Fig. 4. Lavender oil calibration curve using linalool as standard.

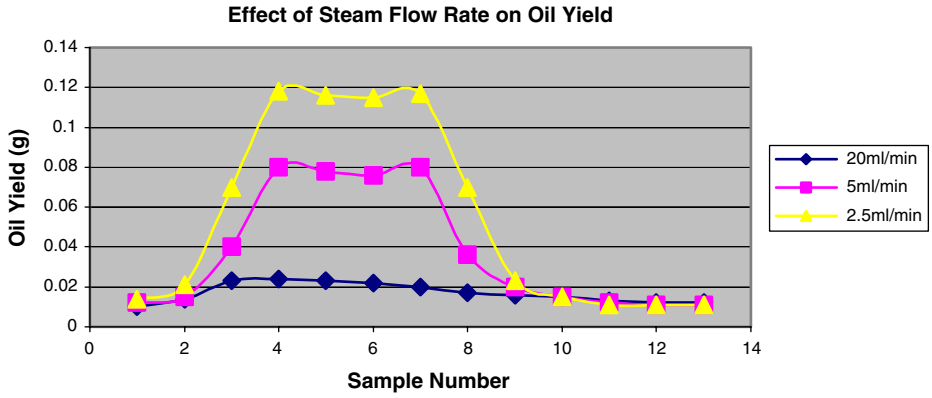


Fig. 5. Artemisia oil sample yield curves at different steam flow rates.

The total oil is the mass equivalent of the principal component in each case.

Distillate samples were collected for analysis to quantify the oil yield in 20 ml samples at different steam and distillate flow rate. The samples were collected during the entire distillation time at equal time intervals. The maximum oil extraction occurred when the steam had the longest residence time in the packed bed. The total oil isolated is the sum of the oil in the oil fraction plus the oil in the aqueous fraction of the 20 ml samples of distillate/condensate.

### 3.1. Oil yield curves

The oil in the oil fraction is given as the oil yield curves in Figs. 5–8.

### 3.2. Pilot scale oil yield

An industrial scale pilot plant was built in Chimanimani in the Eastern Highlands in Zimbabwe for the production of essential oils. It was commissioned and tested based on the experimental work and the newly developed still design. The batches of vegetable samples used were 75 kg. The process generated the average steam or condensate volumes in 25 min. The design will be used to build distillation plants for small businesses interested in cleaner and more economical production of essential oils (Figs. 1 and 2).

In the chromatograms shown in Figs. 9 and 10 the peaks show the different water-soluble compounds in each of the oils of lavender and artemisia. The biggest peak in Fig. 9 is camphor and it was identified by spiking or internal standardisation using the standards bought from Sigma Aldrich Chemicals. The main peak in Fig. 10 is due to linalool, again identified by spiking using a standard linalool sample. The area of each peak was given as a percentage of the total area and that is how the mass of each component can be calculated and the total mass is calculated using Eq. (1).

## 4. Discussion

The optimum insulation thickness on the exterior circumference of the packed bed column has to be calculated so that it is exact. This eliminates excessive condensation in the packed bed. Consequently, the packed biomass bed operates as an isothermal system i.e. a constant temperature column. The high temperature

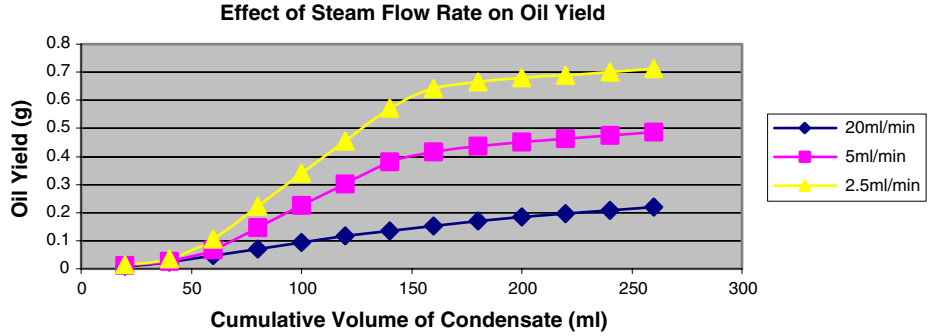


Fig. 6. Artemisia oil cumulative yield curves at different steam flow rates.

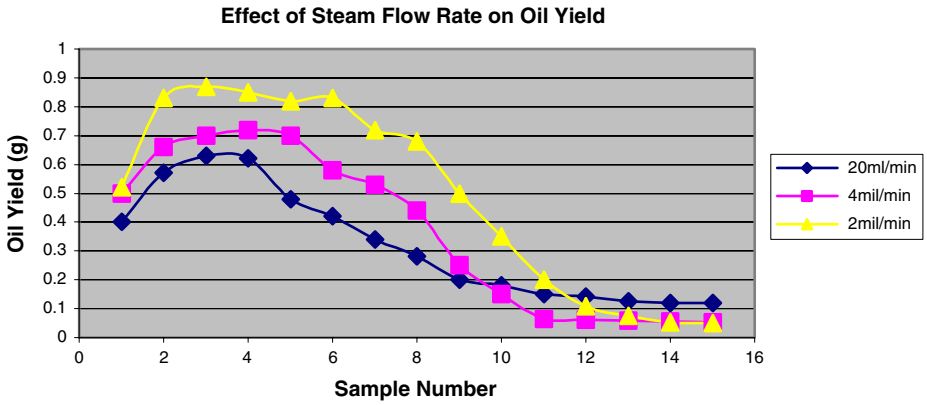


Fig. 7. Lavender oil sample yield curves at different steam flow rates.

that is maintained during the entire distillation period ensures that the water retained in the vegetable mass in the packed column is that which is lost when the steam loses its enthalpy of vaporisation during heat conduction through the biomass. With reduction in heat energy losses to the environment, the packed biomass rapidly reaches the maximum and uniform operating temperature of about 99 °C. The steam jacket ensures distillation at constant elevated temperature. In hydrodistillation, the experimental set up used here would have produced at least 2.9 l of wastewater compared to less than half a litre of wastewater produced in each of these experiments. Wastewater in steam distillation, because of the polar components dissolved in it, is associated with odour nuisance, a form of air pollution, as stated in the book by Jakson [9]. Its reduction increases recovery of oil by increased coalescence rates as well as reduction in unwanted production odours. The oil content in the wastewater averaged 0.24% in artemisia wastewater and 0.26% in the lavender wastewater. For commercial scale distillation plants these values can mean a large loss of valuable product.

The use of a packed bed means that the loss of these water-soluble components can only take place in the aqueous phase of the distillate. In other applications where water and vegetable materials are mixed and brought to a boil, the water in the still retains some dissolved oil, which dramatically increases oil loss. It also causes environmental nuisance because that oil produces odour when it degrades. Using the packed bed column in the distillation means that such losses are eliminated completely and that maximises oil yield and economic benefits in this small-scale industry.

Figs. 5–8 show that it is now possible using the methods outlined here to reduce energy costs in steam distillation operations with increased oil yield. The reason why oil yield improves by as much as two times when steam flow rate is reduced is due to the fact that steam in steam distillation has two uses: to provide heat energy and as a transport medium for the distillate. The marked savings in energy costs and the attendant environmental benefits are illustrated in the yield graphs.

The advantage of this method of using only one component for the analysis of the oil loss to water is that

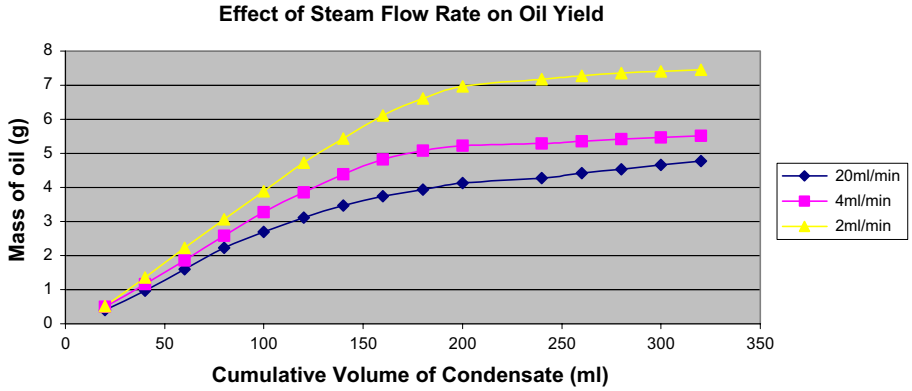


Fig. 8. Lavender oil cumulative yield curves at different steam flow rates.

Table 1
Artemisia oil production

| Condensate volume in old still design without steam jacket (l) | Oil separated (g) | Condensate volume in new still design with steam jacket (l) | Oil separated (g) |
|---|---|---|---|
| 50 | 36 | 30 | 70 |

Table 2
Lavender oil production

| Condensate volume in old still design without steam jacket (l) | Oil separated (g) | Condensate volume in new still design with steam jacket (l) | Oil separated (g) |
|---|---|---|---|
| 52 | 750 | 35 | 480 |

the component being tested for in each type of oil is one of the most valuable components in that oil. This is important given the fact that some essential oils' prices are determined by the percentage composition of some of the components; in this case camphor is highly priced in pharmaceutical applications and so is linalool in fragrances. This type of analysis to monitor purity and/or losses to the water is very important for quality control and loss control. Another advantage of the use of this method is the fact that if one were to seek to analyse for each and every component present in each oil, it would mean analysing for 100 components in the case of lavender and 74 components in the case of artemisia oil; such an exercise would be very expensive. Obtaining standards for each of the components in each type of oil would also be extremely expensive.

## 5. Conclusions

The adoption of the new still with steam jacket design and operation of steam distillation processes result in improved recovery of valuable essential oils (Tables 1 and 2), energy saving and environmental improvement. The steam jacket design of the packed bed column provides a steady-high-temperature distillation process. Towards the end of the distillation cycle the yield drops so low that it is uneconomical to continue distilling. At that stage, relatively more oil is lost to the aqueous fraction of the distillate or condensate than is captured. Using the yield graphs included in this document as a guide, it is easy to determine the cut off point because steam costs continue to be the same while the oil yield drops markedly. The new packed column design reduces

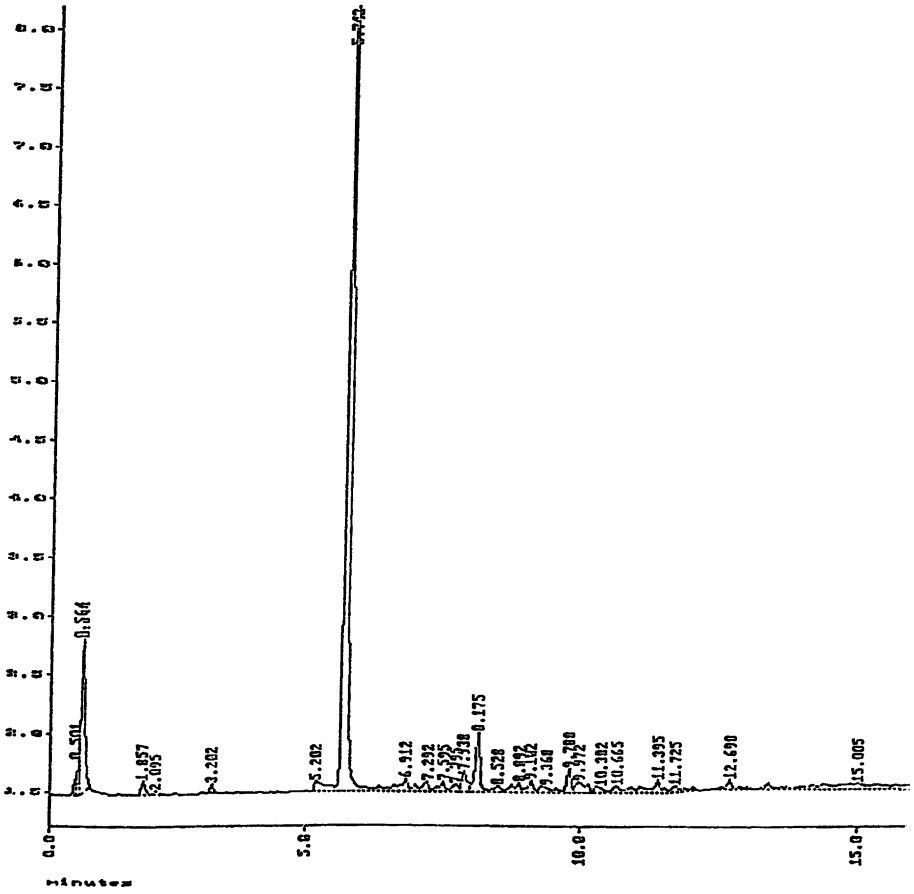


Fig. 9. Chromatogram of water-soluble components in artemisia oil.

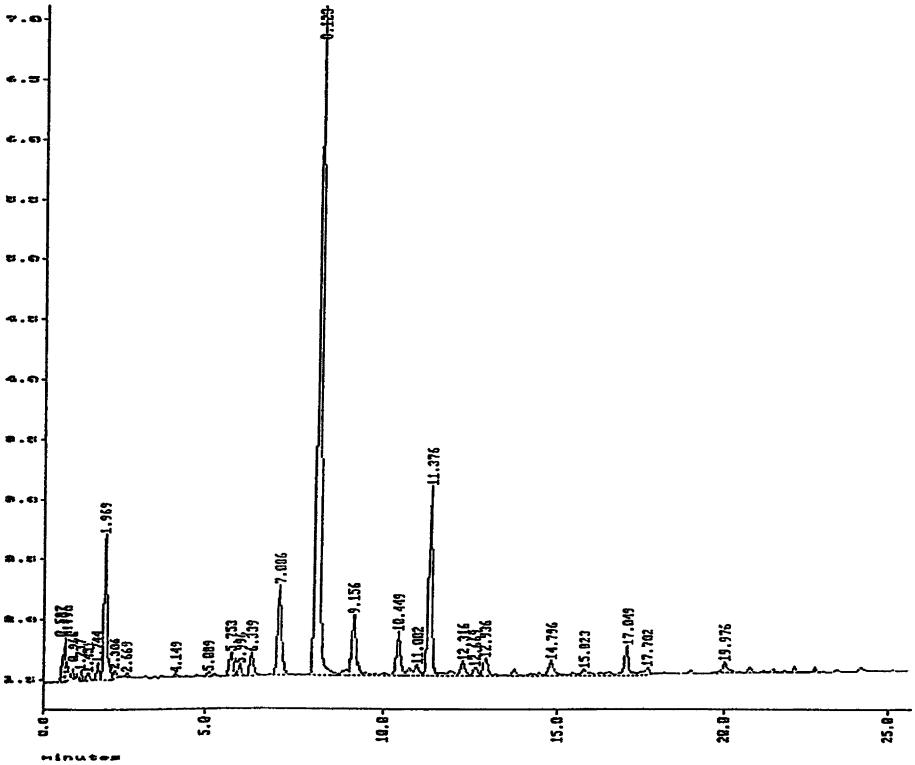


Fig. 10. Chromatogram of water-soluble components in lavender oil.

oil losses considerably when compared with other distillation configurations. The odour nuisance is eliminated or reduced. The odours were completely eliminated in the pilot plant that was used to develop and test the packed bed for the distillation of essential oils. The presence of organic compounds in the wastewater increases the wastewater's chemical oxygen demand (COD), a measure and indication of pollution in water [9]. The principal components in wastewater are camphor in artemisia oil and linalool in lavender oil, both of which are commercially valuable compounds. The results of the pilot scale industrial plant in Zimbabwe document the dramatic improvement in recovered oil products, the savings in energy and very substantial reduction of organics in the wastewater (condensate). The solid residue can easily be utilised via composting.

What is evident here is that by using cleaner production technologies/approaches, the improved product recovery and higher product yield with reductions in environmental burden are win-win benefits for business and the environment.

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