

Research note

Application of Multistage Steam Distillation Column for Extraction of Essential Oil of *Rosemarinuse officinalis L.*

M. Malekydozzadeh¹, P. Khadiv-Parsi^{1*}, Sh. Rezazadeh², H. Abolghasemi¹, Z. Salehi¹, Q. Li³

1- School of Chemical Engineering, College of Engineering, University of Tehran

2- Institute of Medicinal Plant, ACECR, Tehran, Iran

3- School of Chemical Engineering, Curtin University of Technology, Perth, Australia

Abstract

In this work, a new process design and clean production to increase the yield of steam distillation of rosemary essential oil was studied. For extraction of essential oil, a packed bed of rosemary was exposed to the steam flow. For improving the total yield, multistage bed column was used. The effects of steam flow rate, height of plant bed, multistage beds and time of extraction on yield and composition of the oil were studied. Also, trend of three major components of the oil during different extraction times and conditions were observed. We determined minimum steam to be passed through the packed bed so that there is minimum water in the distillation and hence, steam channeling phenomenon was overcome. Furthermore, we obtained optimum steam flow rate and time for extraction of the oil. Using a multistage column with suitable steam redistributors results in reduction of pressure drop, and steam channeling in the column, hence, increasing the yield of extraction. This research revealed different behaviors of the main components of rosemary oil; α -pinene, 1,8-cineole and camphor in treating with amount of steam flow rate.

Keywords: *Multistage Column, Rosemary Plant, Essential Oil, Steam Distillation*

1. Introduction

Essential oils of plants have many applications in medicines, cosmetics, food stuff and pharmaceutical industries. They are present in plants at low concentration which would require high performance extraction techniques in order to achieve high yield. Generally, essential oils are produced by different methods, including solvent extraction, supercritical fluid extraction,

hydro-distillation, steam distillation, use of superheated steam and combinations of the previous techniques with others such as ultrasound and microwave-assisted processes [1,2].

The steam distillation is one of the most popular methods because of its low cost in comparison with advanced methods such as supercritical fluid extraction and its green approach compared to solvent extraction.

* Corresponding author: kparisi@ut.ac.ir

This method has been actively pursued since the beginning of the 1980s. In the literature there are some studies of oil extraction by steam distillation [3]. Masango studied the effects of increasing the steam flow rate and steam jacket as well as appropriate insulation of distillation column in extracting essential oil from Artemisia and Lavender plants by steam distillation, optimizing the consumption of energy and increasing the final yield of extraction [2]. Romdhane et al. presented a mathematical model for optimizing the steam distillation process for preparation of Pimpinella anisum oil [4]. Using a microwave heater instead of an electric one decreased the time of extraction and improved the quality of Artemisia oil [1]. In another study the effect of crushing of the plant and time of extraction on the yield and chemical composition of Coriander oil was reported [5]. Rosemary (*Rosemarinus officinalis* L.) is an aromatic, medicinal and condiment plant that belongs to the family labiatae, reaching a height of 1.5 meters. Essential oil of rosemary, known as rosemary oil, is obtained by steam distillation method of the fresh leaves and twigs. The yield ranges from 0.5 to 1.5 % (w/w) [6]. Chemical composition and physicochemical characteristics have been reported for rosemary essential oil. It is an almost colorless to pale yellow liquid with a characteristic, refreshing and pleasant odor. Major components characterized for the oil are α -pinene, 1,8-cineole and camphor [7]. The effect of extraction time on the yield and composition of rosemary oil has been reported in two different methods; steam distillation and hydro-distillation [2]. The composition of oil may vary to a large

extent depending on the extraction method used. Steam distillation process was modeled as an inevitable step to project industrial plants with good operational condition [8]. Bimakr et al. showed that conventional soxhlet extraction of flavonoids of spearmint took 6 hr for 3 g of dried and ground plant at 40°C using solvents such as ethanol, methanol. So, they proposed supercritical extraction with CO₂ to obtain high efficiency [9]. Therefore, solvent extraction does not perform well.

In the present study, a column was designed and constructed in order to evaluate the steam distillation process for the extraction of rosemary oil. Optimal operating conditions were determined in order to achieve high extraction yields. Also, we analyzed the effect of different parameters on chemical composition of extracted rosemary oil. The novelty of the process is introducing multistage beds of plant in order to achieve a better extraction of target materials through enhancing mass transfer operation.

2. Material and method

2-1. Plant material

Rosemary plant was collected (December 2006) from the Iranian Institute of Medicinal Plants (IMP) research farm and dried on a bench in the shade.

2-2. Steam distillation apparatus and procedure

A schematic diagram of the steam distillation apparatus used for essential oil extraction is shown in Fig. 1.

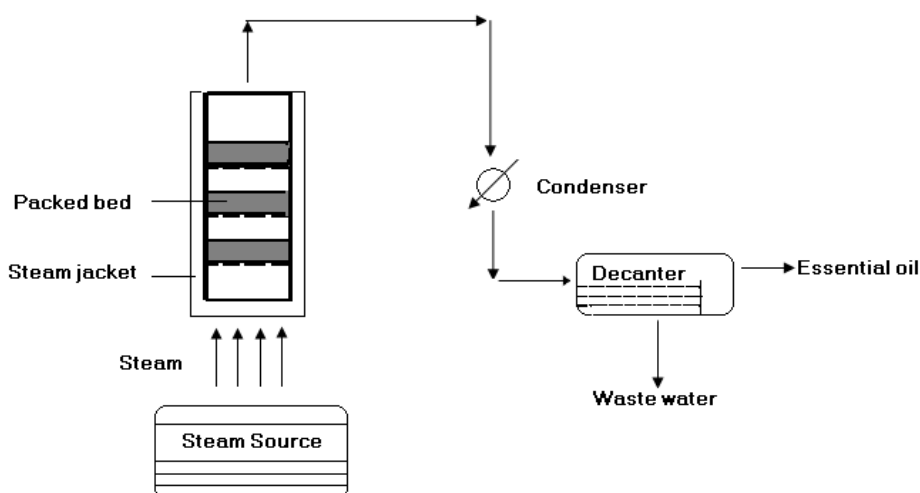


Figure 1. Schematic diagram of the steam distillation apparatus

The apparatus has a cylindrical Pyrex body (6 cm inside diameter and 60 cm height). A batch of 100-200 g of dried and ground leaves of rosemary was packed in the column with 2000 ml water in steam source. The raw material forms the packed bed. The lid was closed and the process of distillation began with the injection of steam to the bottom of the column. Each plant bed was exposed to several flow rates of steam. Steam and essential oil were condensed and collected in time intervals of 5, 15, 30, 60 and 100 minutes. Following condensation, the mixture was decanted to separate phases of oil and water. The essential oil was collected, dried with anhydrous sodium sulfate and stored at 4°C until analyzed.

The essential oil yield was estimated according to the dry vegetal matter by using the following equation [7]:

$$Y = \frac{M_{oil}}{M_s} \times 100 \quad (1)$$

Where:

M_{oil} : Mass of essential oil [g].

M_s : Mass of dry plant [g].

Y : Essential oil yield [w/w].

In these experiments, the steam jacket of the column was insulated by foam cover having 1 cm thickness.

After doing the experiments and determining the yield of each experiment, the obtained samples from the three experiments with steam flow rates of 4, 7 and 9 l/min and a packed bed of 100 g that were collected at five intervals, namely 5, 15, 30, 60 and 100 minutes, were analyzed by GC-MS and GC instruments. Each experiment was repeated at least three times and mean of results was reported.

2-3. Gas chromatography-mass spectrometry identification

GC analyses were carried out using a Hewlett-Packard 6890 with HP-5 capillary column (phenyl methyl siloxane, 25 m×0.25

mm, 0.25 μm film thickness) and a DB-1 capillary column (30 m \times 0.25 mm, 0.25 μm film thickness). The temperature of oven was programmed to 60–240°C at 4°C/min; injector temperature, 250°C; detector temperature, 260 °C; carrier gas, He (1.5 ml/min); split ratio, 1:25. GC–MS analyses were carried out applying a Hewlett-Packard 6859 with a quadropol detector, on a HP-5 column (see GC), operating at 70 eV ionization energy, using the same temperature programmer and carrier gas as above.

3. Results

The isolation and concentration of essential oils were performed in single stage and multistage column with three steam flow rates. The yield was calculated from the relation between the mass of obtained oil and the mass of raw material used in the experiments.

The three different steam flow rates (4, 7 and 9 l/min) and a packed bed of 100 g were used. The results are shown in Fig. 2.

It can be seen from this figure that as steam flow rate decreases, the amount of oil increases monotonically in each of the five intervals. The greatest yield was obtained for the steam flow rate of 4 l/min, which was 1.074. Also, from the sharpness of the slope during the first 30 minutes, we observe that the highest extraction rate occurs in the first 30 minutes of extraction time. In general, in most cases of the experiments, between 85 and 95 percent of total oil was extracted during this interval.

The effect of height of packed bed on the extraction yield of essential oil was considered for two steam flow rates of 4 and 7 l/min. and packed beds with the heights of 30, 45 and 60 cm. These heights were equal to 100, 150 and 200 g of plant, respectively. The results of these experiments are presented in Figs. 3 and 4. It was evident from these experiments that with an increase in the mass of the packed bed, a sharp decrease occurred for the total yield of the extraction, depending on the steam flow rate.

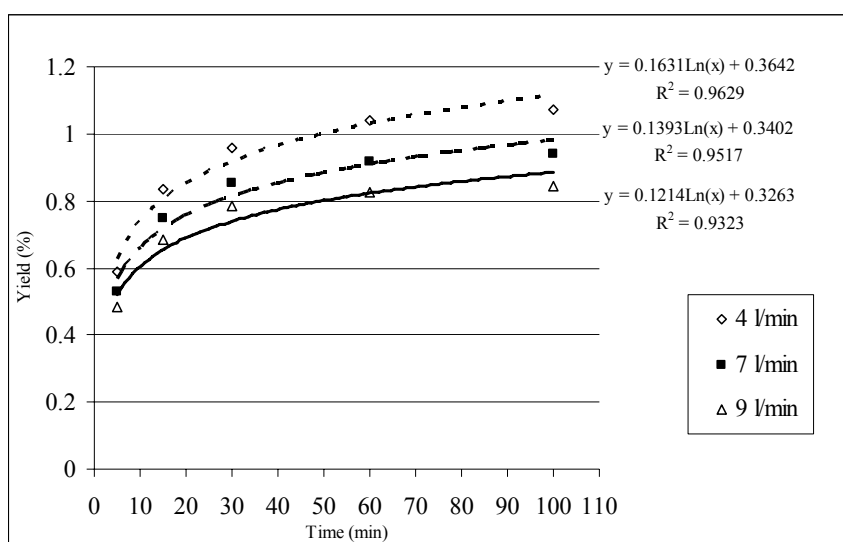


Figure 2. Extraction yield as a function of processing time under different steam flow rates.

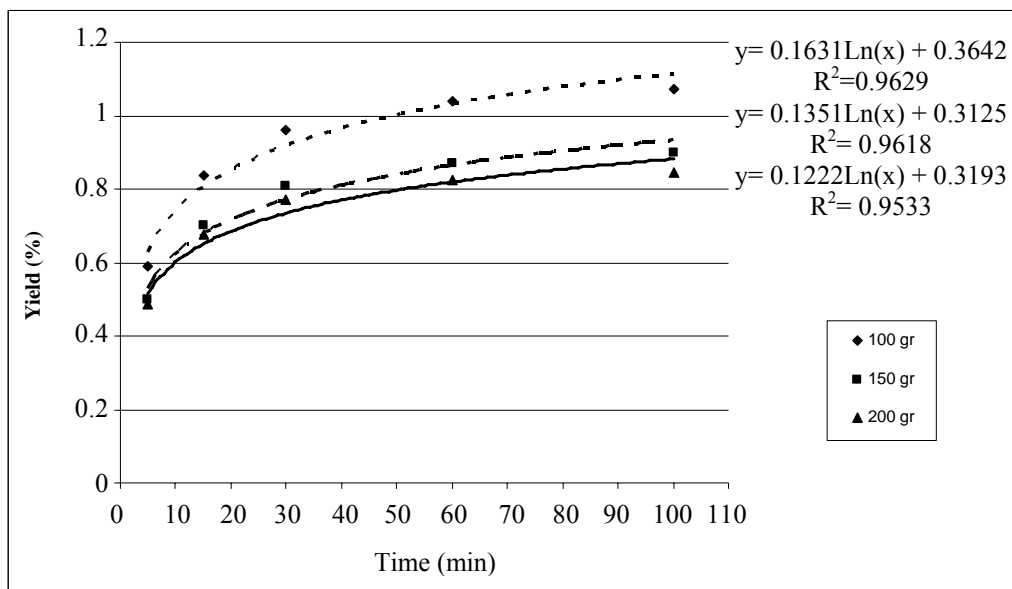


Figure 3. Yield curves of rosemary oil samples for different masses of packed-beds for steam flow rate of 4 l/min.

Furthermore, the slope of the curves for the packed beds of 45 and 60 cm of height were less during the 30 minutes in comparison with that of 30 cm height of bed. It revealed

that the amount of extraction was less in this interval for higher heights of single packed bed.

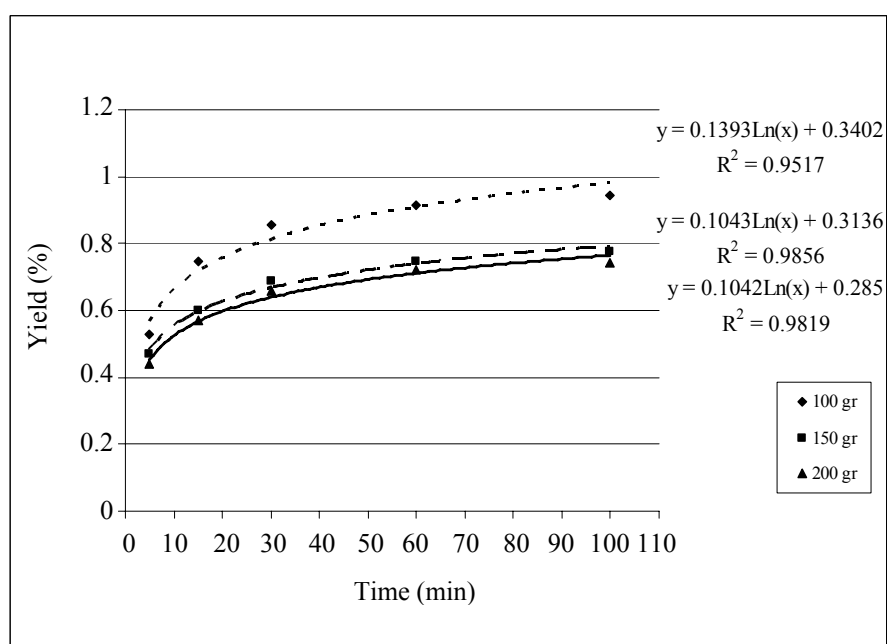


Figure 4. Yield curves of rosemary oil samples for different masses of packed beds for steam flow rate of 7 l/min.

An increase in the height of the packed bed caused higher pressure drop. As a result, the total yield of the extraction decreased. To avoid this problem, steam redistributors were used between every two successive beds of the column.

As seen in Figs. 5 to 7, using two or three stages of beds caused an increase in the total yield of the extraction, and the yield of such process in one-stage column with a mass of 150 g and a steam flow rate of 7 l/min reached from 0.776 percent to 1.03 percent in a three stage column with the same condition (Fig. 5).

As shown in Fig. 6, reduction of total mass of beds at the same steam flow rate of the

previous condition enhanced the yield.

With respect to Fig. 7, a decrease in the steam flow rate and also a decrease in the mass of the packed plant with the same height of column caused a yield increment up to 1.36 percent.

However, using more stages does not cause any increase in the total yield. With a decrease in the height of the packed bed, a decrease in the steam channelling was observed. But, here the contact between steam and the internal walls of the column increased, which in turn caused the steam condensation on the internal walls of the distillation column which returned to the packed bed.

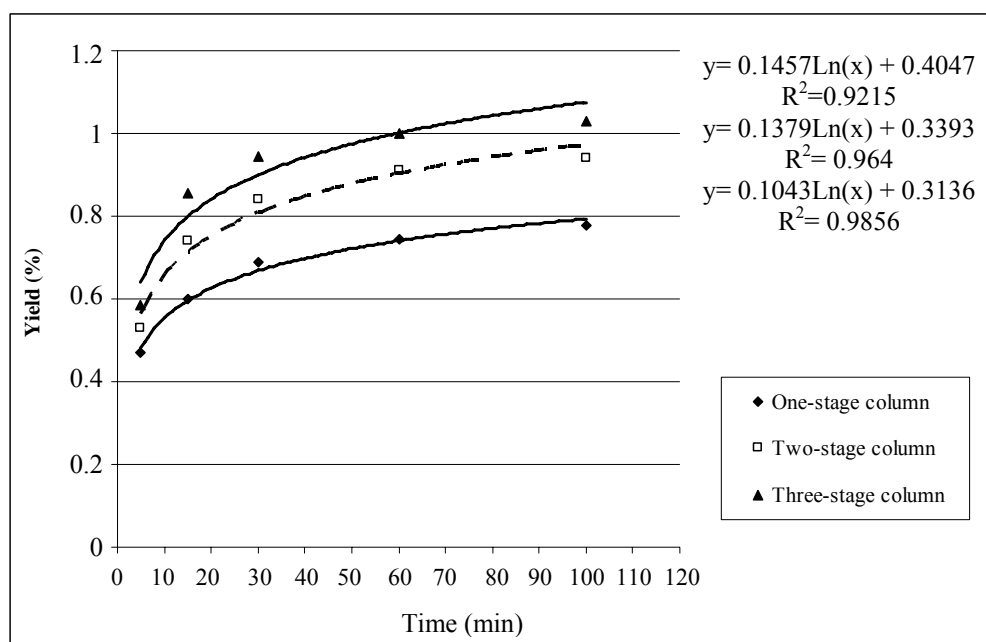


Figure 5. Yield curves of rosemary oil samples for multistage column with steam flow rate of 7 l/min and packed bed of 150 g.

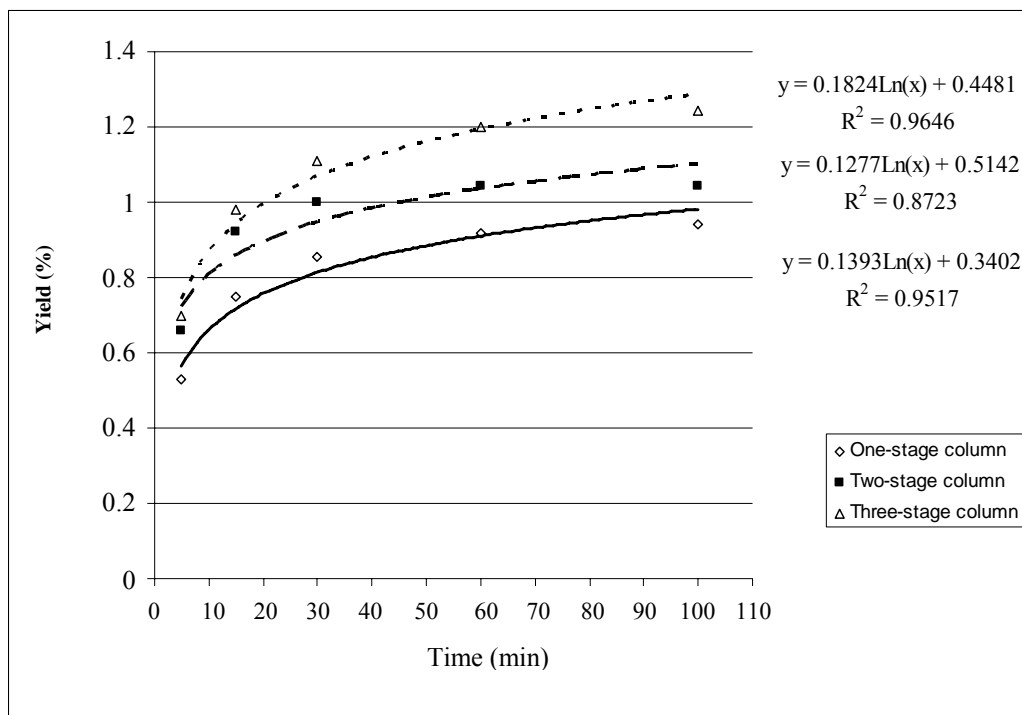


Figure 6. Yield curves of rosemary oil samples for multistage column with steam flow rate of 7 l/min and packed bed of 100 g.

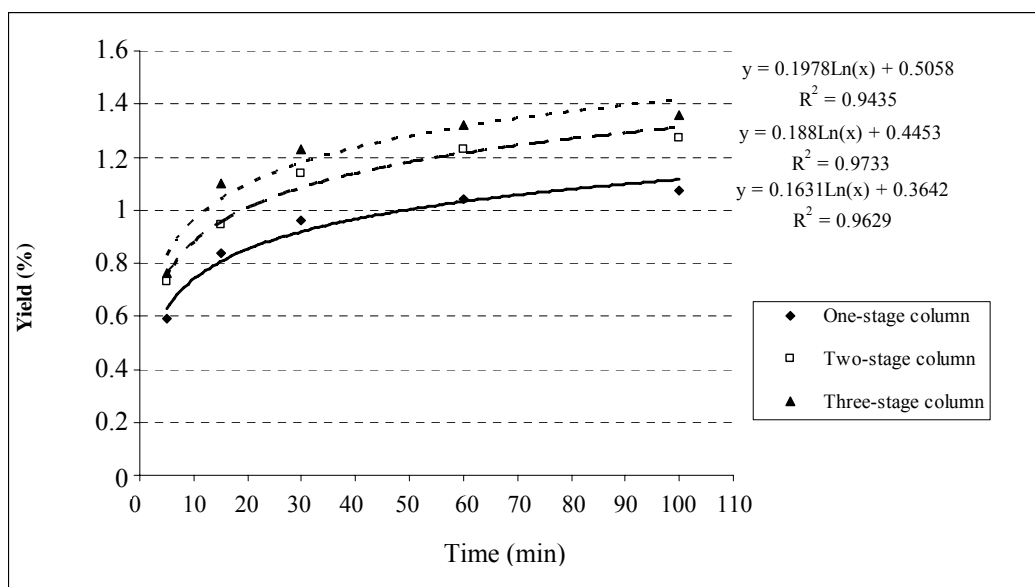


Figure 7. Yield curves of rosemary oil samples for multistage column with steam flow rate of 4 l/min and packed bed of 100 g.

The results of GC-MS analysis showed that rosemary oil has 61 components with the concentration between 0.01 and 15.47

percent. In Table 1 the major components of the rosemary oil are presented.

Table 1. The important components of essential oil of rosemary.

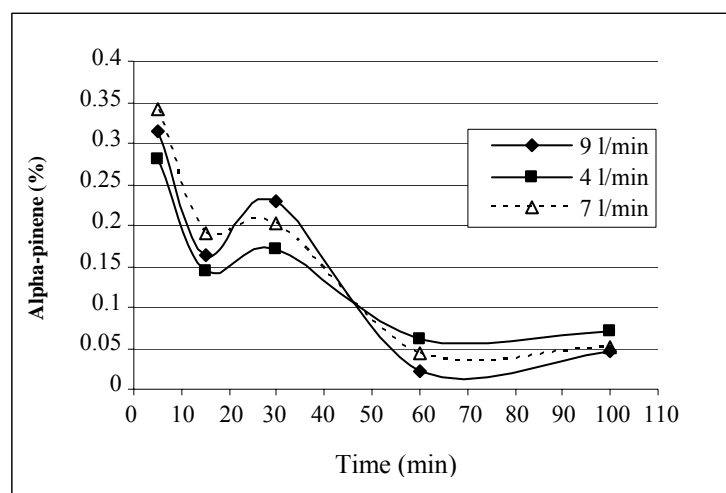
No.	Components	Retention time (min)	Concentration (%)
1	α -pinene	16.78	15.47
2	Camphene	17.60	6.01
3	3-octanone	19.63	5.95
4	Myrcene	19.82	3.92
5	Limonene	21.87	2.26
6	1,8-cineole	22.26	12.14
7	Linalool	25.83	4.51
8	Camphor	28.43	10.04
9	endo-borneol	29.56	7.25
10	α -terpineol	30.73	2.49
11	Bicycle [3.1.1] hept-3-en-2-one	31.81	8.55
12	bornyl-acetate	35.04	2.60

The results of gas chromatography were analyzed for three experiments with steam flow rates of 4, 7 and 9 l/min and a packed bed with a mass of 100 g. The three most important components (α -pinene, 1,8-cineole, and camphor) were selected and changes in their concentration in relation to the time of extraction were studied during 5, 15, 30, 60 and 100 min intervals (Figs. 8 to 10).

For α -pinene a maximum extraction occurred in the first 5 minutes, following a decrease

during 15 minutes and an increase in the further 30 minutes of extraction was observed.

For 1,8-cineole, decrease in extraction amount occurred during the time of extraction. Low extraction was observed for camphor in the first 5 minutes; a maximum extraction occurred at 15 minutes and was followed by decreasing with time. Varying steam flow rate had no effect on the behavior of each component.

**Figure 8.** Change of α -pinene concentration in terms of time for different steam flow rates.

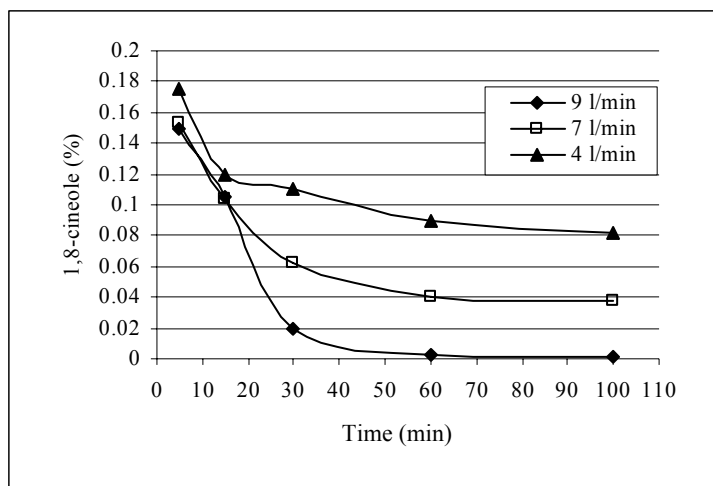


Figure 9. Change of 1,8-cineole concentration in terms of time for different steam flow rates.

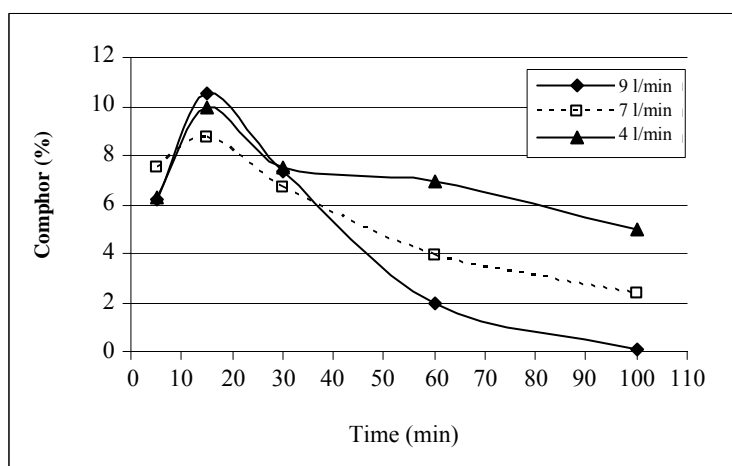


Figure 10. Change of camphor concentration in terms of time for different steam flow rates.

4. Discussion

The configuration of the system used in the present work was effective to extract the essential oil of rosemary. The experiments of steam distillation that were conducted proved that extraction yield of rosemary decreased with steam flow rate. Decrease in the steam flow rates increases the time of steam lingering in separation process. Consequently, the contact time between steam and plant tissue increases, this will provide enough opportunity for essential oils to evaporate and be extracted. A decrease in

the steam flow rate will affect the extraction process and has an economic impact due to reduced energy consumption.

Experiments that were conducted in multistage columns proved that an increase in the number of stages has a considerable effect upon yield up to 2 percent. However, this factor decreases for the column with more than three stages; since increasing the number of stages will result in the increase in steam contact with walls of the column and will cause steam condensation on it.

An increase in height of the bed inside the separation column causes a decrease in the process yield, since an increase in the height of the bed leads to an increase in pressure drop across the bed of plant. As a result a fast evaporation occurs. Increasing the evaporation rate creates the phenomenon of channeling inside the bed. In other words, steam moves towards the walls of the column and the contact surface between the steam beds of the plant decreases.

Experiments showed variations in the amount of components with time. The results indicated weak effect of steam flow rate on the essential oil components. In other words, a change in the amount of steam flow rate does not lead to either increase or decrease in any considerable components of Rosemary essential oil. However, the time of distillation has an effect on the constituents of essential oil. This effect was studied for three important components of Rosemary essential oil: α -pinene, 1,8-cineole, and camphor. Each of these components showed different behavior in contact with steam flow rate during time intervals. First, barrier effect, and second, the boiling points of components. In the case of α -pinene, barrier effect is active and for camphor and 1,8-cineole, the boiling point is effective. It should be noted that some of the α -pinene is present in the cell of plant walls and some exists inside these cells. This increase and decrease is due to evaporation of compounds that exist between these cells. Another reason is the time needed to destroy the cell walls that contain essential oils. But in the case of camphor and 1,8-cineole, this trend is the result of their boiling points. 1,8-cineole has the boiling point of 176 °C, and camphor has

the boiling point of 209 °C. Each of these components showed different phenomenon in contact with steam flow rate during time intervals.

Cassel et al. [8] used steam distillation for wet leaves. The steam distillation apparatus used includes a 4.0 ml glass boiler and a 3.0 ml glass extraction chamber. The steam flow rate was 3.4 ml/min for 225.57 g of rosemary. GC-MS of essential oil showed 37.22% of α -pinene, 23.76% of 1,8-cineole and 0.4% camphor. Roldan-Guiterrez et al. [10] postulated that extraction time in the method of ultrasound-assisted dynamic extraction of valuable compounds from plants and flowers such as rosemary was 165-176 minutes less than steam distillation, depending on the target compound. Rezzoug et al., used fast controlled pressure drop following steam distillation using 100 g of rosemary leaves. They showed that optimum processing time for α -pinene and 1,8-cineole was 10.4 minutes at 309 kPa [11]. Tigrine-Kordjani et al. [12] used solvent free microwave extraction for 200 g of fresh rosemary with a fixed power of 200 W for 30 minutes. They showed that geographical origins of rosemary influenced the percent of target constituents. The difference between the present research and the results mentioned above is due to application of multistage bed in distillation column.

5. Conclusions

To the best of our knowledge, multistage column has rarely been used in a steam distillation device. This method brings a good result concerning efficiency in rosemary oil extraction. Also, the maximum oil extraction occurred when the steam had the longest residence time in the packed bed, because steam had enough time to destroy the plant cell walls. Also, the finely divided

plant had better results. Therefore, it would be applied in industrial cases. This study made it possible to identify the phenomenon of some important components of the rosemary oil during steam distillation. In addition, the results showed that the duration of extraction should be at least 30 minutes, because more than 85% of the oil is extracted at the end of this time. This approach provides a cost-effective and clean production route for plant oil extraction.

References

- [1] Chemat, F., Lucchesi, M., Smadja, J., Favretto, L., Colnaghi, G., and Visinoni, F., "Microwave accelerated steam distillation of essential oil from lavender: A rapid, clean and environmentally friendly approach", *Analytica Chimica Acta*, 555, 157 (2006).
- [2] Masango, P., "Cleaner production of essential oils by steam distillation", *J. Cleaner Production*, 13, 833 (2005).
- [3] Rajeswara, Rao, B. R., Kaul, P. N., Syamasundal, K. V. and Ramash, S., "Water soluble fractions of rose-scented geranium (pelargonium species) essential oil", *Bioresource Technology*, 84,243 (2002).
- [4] Romdhane, M. and Tizaoui, C., "The kinetic modelling of a steam distillation unit for the extraction of aniseed (pimpinella anisum) essential oil", *J. Chem. Technol. Biotechnol.*, 80,759 (2005).
- [5] Smallfield, B. M., Van Klink, J. W., Perry, N. B. and Dodds, K. G., "Coriander spice oil: Effects of fruit crushing and distillation time on yield and composition", *J. Agric. Food Chem.*, 49, 118 (2001).
- [6] Mateus, E. M., Lopes, C., Nogueira, T., Lourenco, J. A. A. and Marcelo Curto, M. J., "Pilot steam distillation of rosemary (*Rosmarinus officinalis L.*) from Portugal", *Silva Lusitana*, 14, 203 (2006).
- [7] Boutekedjiret, C., Bentahar, F., Belabbes, R. and Bessiere, J., "Extraction of rosemary essential oil by steam distillation and hydrodistillation", *Flavour and Fragrance J.*, 18, 481 (2003).
- [8] Cassel, E., Vargas, R. M. E., Martinez, Lorenzo, N. D. and Dellacassa, E., "Steam distillation modeling for essential oil extraction process", *Industrial Crops and Products*, 29, 171 (2009).
- [9] Bimakr, M., Rahman, R.A., Saleena, F. S., Ganjloo, A., Md Salleh, L., Selamat, J., Hamid, A. and Zaidul, I. S. M., "Comparison of different extraction methods for the extraction of major bioactive flavnoid compounds from spearmint (*Mentha Spicata L.*) leaves", *Food and Bioproduct Processing*, 89, 67 (2011).
- [10] Roldan-Gutierrez, J. M., Ruiz-Jimenez, J. and Luque de Castro, M. D., "Ultrasound-assisted dynamic extraction of valuable compounds from aromatic plants and flowers as compared with steam distillation and superheated liquid extraction", *Talanta*, 75,1369 (2008).
- [11] Rezzoug, S. A., Boutekedjiret, C. and Allaf, K., "Optimization of operating conditions of rosemary essential oil extraction by a fast controlled pressure drop process using response surface methodology", *J. Food Eng.*, 71, 9 (2005).
- [12] Tigrine-Kordjani, N., Chemat, F., Meklati, B.Y., Tuduri, L., Giraudel, J. L. and Montury, M., "Relative characterization of rosemary samples according to their geographical origins using micro-wave-accelerated distillation, solid-phase microextraction and Kohonen self-organizing maps, *Anal. Bioanal. Chem.*, 389,631 (2007).