Evaluation of Influencing Factors and Technological Processes for the Production of Orange Essential Oil (*Citrus sinensis*) on Equipment Semi-industrial Distillation

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Abstract: *In recent years, essential oils extracted from different plant species have become increasingly popular in the production of pharmaceuticals, cosmetics, and foods. The essential oil from orange (*Citrus sinensis*) is important in large-scale applications due to its antibacterial, antioxidant activities, and pleasant aroma. In this study, factors affecting the production of orange essential oil on a distillation device with an operating capacity of 50 L/batch, including the ratio of material to water, temperature, and time distillation, have been surveyed. Through the survey, it was found that the raw materials were pureed, the materials: water ratio was 1:3 g/g, the water heating temperature was 130 °C, and the distillation time was 140 min. The yield of the essential oil was 1.8 mL/g with compounds limonene accounting for 98%, α-Pinene (0.655–0.734%), and β-Pinene (1.114 and 1.163%) by the GC-MS method. The review also found that the hydrodistillation equipment was designed to be suitable for the semi-industrial scales of orange essential oil due to its stable yield and volatile compounds contained in the essential oil.*

Keywords: Citrus sinensis*; hydrodistillation; limonene; essential oil*

INTRODUCTION

Aromatic substances in general or essential oils have been associated with human life and civilization for thousands of years. Essential oils are the very small, quintessential, most concentrated drops of herbs. Sometimes it's just aromatic molecules formed through regulatory functions in the stems of plants that make them attractive, seduce, bring vitality, freshness, and bring richness to life [1-3]. It is a mixture of soluble organic substances with a characteristic odor. Essential oils are widely used in the cosmetic industry, especially in the production of various perfumes [4], shower gels, hair creams, shampoos [5], as a component of disinfectants in pharmaceuticals [6], and as pesticides in the field of insecticides from natural substances [7-8]. Phenolic constituents, which are present in essential oils, are known to have antibacterial activity, and some are generally recognized as safe substances [9-11]. Therefore, it is used to prevent the growth of bacteria and contaminants in food [6,12-13].

In recent years, the use of herbs in production in many fields has become more and more popular. Various studies illustrate the activities and effectiveness of compounds derived from medicinal plants. Orange tree (*Citrus sinensis*) is grown mainly in different Asian countries such as Vietnam and Thailand. Orange fruits have great potential for scientific research due to the presence of a large number of bioactive compounds with antibacterial and antioxidant activities [14-17]. Previous research showed that there are different compounds obtained from orange peels essential oil, such as limonene (main compound accounting for over 70%), myrcene, terpinolene, etc., which are among the important components of oranges, help prevent carcinogenic, antibacterial, and aromatherapy properties in physiotherapy [12,18-19]. Orange essential oil (OEO) has an important role in aromatherapy and cosmetic applications due to its antibacterial activities. The quality of OEO was evaluated based on the analytical method GC-MS. Among the extraction methods of essential oils, the hydrodistillation method has been widely used, especially for commercial production, because of its low cost, ease of operation, and efficient recovery stable, the high-quality essential oil obtained [4,20-21].

In Vietnam, the value of orange raw materials changes with great variability from time to time, thus making the value of this product unstable and making life difficult for farmers. Enhancing the economic value of agricultural products is of interest, and the production of essential oils from these agricultural products is a promising option that can enhance the value of these agricultural products. Studies on hydrodistillation methods as well as optimization of technological parameters to recover essential oil from orange peel have been carried out for a long time. However, most of the studies have been done in flasks, at the laboratory scale, with few published studies on the semi-industrial or industrial scale. It's a long way from laboratory-scale research to industrial-scale application. Differences in production scale will significantly affect the yield and quality of essential oils. Pilot-scale studies are needed to make it easy to apply in practice.

The purpose of this study is to investigate the influencing factors and technological process of producing OEO (*Citrus sinensis*) by a 50-L hydrodistillation device. Specifically, surveying the conditions affecting the content of OEO (ratio of water/material, temperature, distillation time); evaluating the chemical composition of the optimized essential oil sample, and the performance of a 50-L hydrodistillation device.

■ EXPERIMENTAL SECTION

Materials

This study was conducted on the subject of oranges (*Citrus sinensis*) in Hau Giang province (9° 45'52" N 105° 38'25" E) harvested in July 2021. Oranges purchased must be fresh, not damaged, have the characteristic green color of oranges, no strange smell. In addition, sodium sulfate (Na_2SO_4, Sigma Aldrich) was used for the anhydrous process of the essential oil after it was obtained.

Instrumentation

Equipment used in the study includes a distillation device with a volume of 50 L (diameter = 300 mm, thickness = 3 mm, height = 650 mm) heated by thermal oil (140–160 °C), imported capacity raw material from 20 to 25 kg/batch (Fig. 1). The equipment is made of stainless steel and inox 304. In addition, the unit is equipped with a horizontal beam tube condensing system cooled by water, a condensate recovery device, temperature control and sensing system, and pressure gauge.

Procedure

Hydrodistillation

After being harvested, the oranges are pre-treated, damaged materials removed, and peeled and flesh separated. The peels were grounded into small pieces and put into the material tank of the device 50 L. The amount of water was added in accordance with each experiment and put into the distillation system at a ratio of 1:2–1:5 (g/g). The heating temperature is at 120–150 °C,

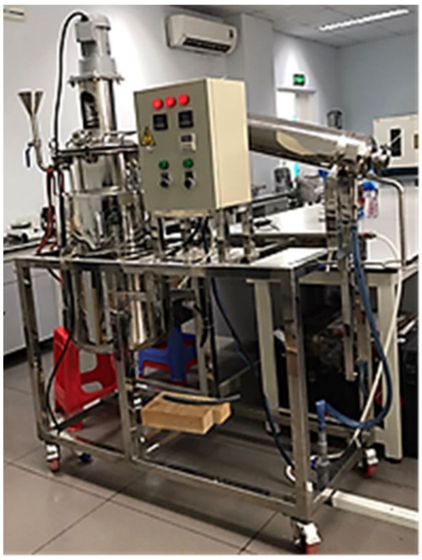


Fig 1. Hydrodistillation equipment 50 L/batch

and the distillation time was calculated from the moment the first drop of liquid appears until the amount of essential oil remains constant. After the essential oil extraction process, a mixture of water and essential oil was obtained after passing through the condenser to recover the essential oil. Then, Na_2SO_4 salt was added to remove the water content in the essential oil. After the water removal process, pure essential oil was obtained.

The yield of essential oil in orange peel is determined as in Eq. (1):

$$Y\left(\frac{mL}{g}\right)=\frac{V}{m}\times100\% \quad (1)$$

where V: volume of obtained essential oil (mL), m: weight of fresh orange peel (g)

Analysis of compounds by Gas Chromatography-Mass Spectrometry (GC-MS)

The chemical composition of the essential oils was determined by GC-MS analysis using the Agilent 6890 N GC instrument in combination with an inert HP5-MS and MS 5973 column. Head column pressure is 9.3 psi. Twenty-five µL of essential oils were added with 1 mL of *n*-hexane and dehydrated with Na_2SO_4. The flow rate is constant at 1 mL/min. Nozzle temperature is 250 °C, and the division rate is 30. Heating program for samples: 50 °C is held for 2 min, increase 2 °C/min to 80 °C, continues to increase 5 °C/min to 150 °C, keep increasing at 10 °C/min to 200 °C, increase 20 °C/min to 300 °C, and hold for 5 min.

Statistical analysis

Each experimental data was presented as mean ± standard deviation in three replicates. Statistical analysis was carried out by using SPSS software (SPSS Inc, Chicago, USA). The analysis of variance (ANOVA) and Tukey HSD analysis were used to compare the significant difference among analyzed groups at the significant level of 5%.

■ RESULTS AND DISCUSSION

Conditions Affecting the Yield of the Orange Essential Oil (OEO)

The influence of the raw materials and water ratio

During the hydrodistillation process, at the boiling point, the essential oils in the plant cells diffuse out to the surface of the material and are carried away by the steam. At the same time, water enters the material in the opposite direction, and the oil continues to be entrained in the water. The process continues until the essential oil in the tissues is completely drained out. Therefore, determining the percentage of water in the raw material is necessary for the extraction process to achieve maximum yield. Experiments were carried out on two-weight scales of orange peels material, 3 kg and 5 kg. The results presented in Fig. 2(a-b) show that when increasing the ratio of raw materials and water, the yield of the obtained essential oil tends to increase to a certain value, and then this yield decreases. Accordingly, at the ratio of 1:2 (g/g), the low amount of essential oil obtained is due to the insufficient amount of water needed to dissolve the colloidal film surrounding the essential oil bag. This leaves a large amount of residual essential oil in the material, reducing the efficiency of the essential oil distillation process. On the other hand, at a higher ratio of 1:5 (g/g), the amount of essential oil obtained is not high because excess water can dissolve or form an oil emulsion and oil loss in the tube collection, reducing the efficiency of the hydrodistillation process. In practical terms, the large volume of water also leads to longer distillation times and higher operating costs. Therefore, the highest yield of essential oil was obtained at 1.63 (mL/g) with a ratio of material:water of 1:3 (g/g). This ratio is also consistent with previous studies on the distillation of Citrus essential oils, such as Tran et al. [13] on the distillation of *Citrus latifolia* essential oil; or by Ngo et al. [22] on the distillation of *Citrus aurantifolia* essential oil.

For the 5 kg weight, the yield of orange oil obtained on the 50 L/batch device did not change significantly compared to the 3 kg weight. Accordingly, the ratio of 1:3 (g/g) still gave the highest yield, accounting for 1.67 (mL/g), followed by the ratio of 1:2 (1.5 mL/g), 1:4 (1.54 mL/g), and 1:5 (1.44 mL/g). Therefore, through the survey results, it was found that with 3 kg and 5 kg of input materials, there was no significant change in the efficiency, and the optimal ratio was still chosen as 1:3 (g/g).

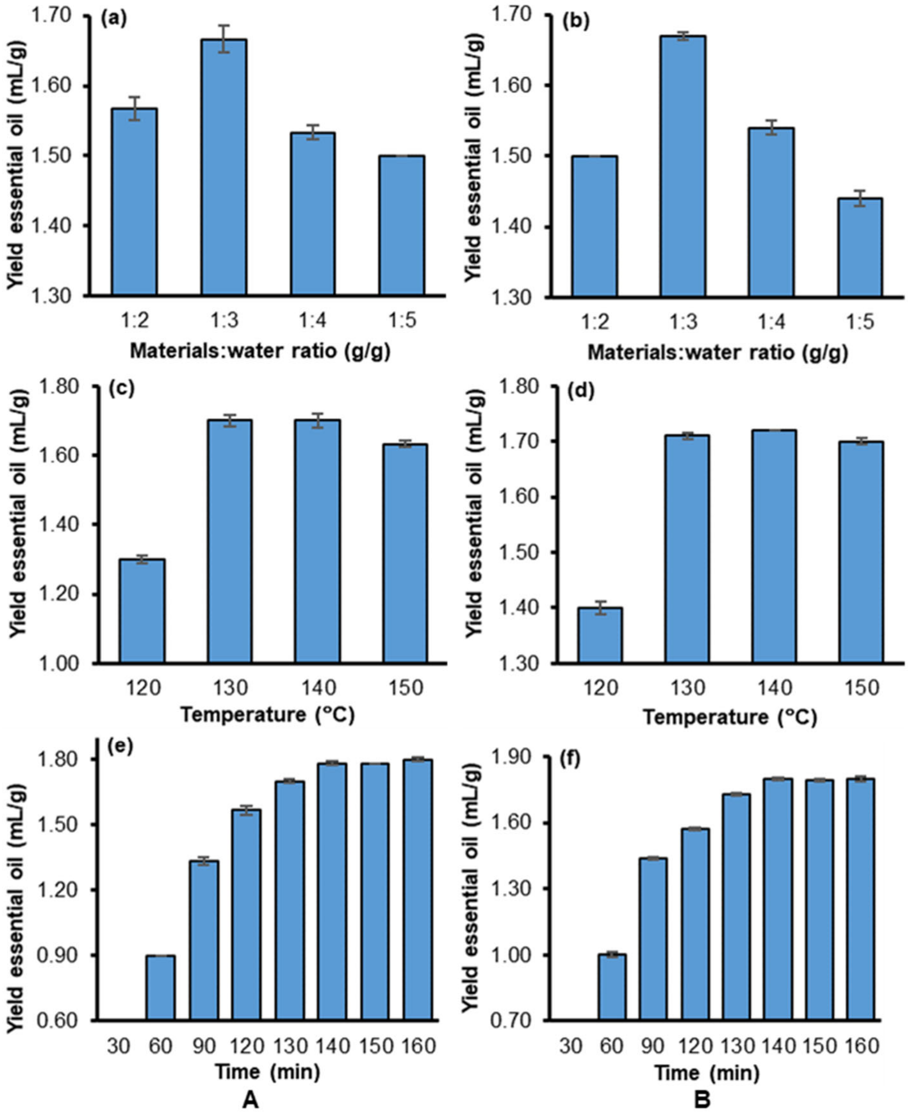


Fig 2. Factors affecting the hydrodistillation of orange essential oil on a 50L/batch device (A. 3kg scales, B. 5kg scales)

The influence of hydrodistillation temperature

One of the main factors affecting the hydrodistillation process is temperature. Fig. 2(c-d) was presented the surveying temperature conditions from 120 to 150 °C, which is consistent with many previous studies on optimizing the extraction process of Citrus essential oils such as Tran et al. [13], Ngo et al. [22], and Phat et al. [23]. The results from Fig. 2(b) show that high temperature leads to better oil yield, but only to a certain point. When the temperature exceeds the optimum point, the yield will decrease afterward. Specifically, in both 3 kg and 5 kg materials, the yield of essential oil increased from 1.3 to 1.63 mL/g (3 kg of material). It increased from 1.4 to 1.7 mL/g (5 kg of material) when the temperature was increased from 120 to 140 °C. However, when analyzing each temperature, at 130 °C, the oil yield reached the highest value of 1.7 mL/g (3 kg) and 1.71 mL/g (5 kg). In contrast, at the temperature of 140–150 °C, the yield of essential oil decreased, accounting for 1.63 and 1.72 mL/g. This can be explained because the high temperature makes the heat transfer and mass transfer from the outside to the material faster due to the weakening of the surface tension and the viscosity of the water. On the other hand, at too high temperature, there are also some limitations. Some components of essential oils are sensitive to decomposition temperature, adversely affecting extraction efficiency, essential oil quality, and production cost due to when the high temperature will increase energy consumption. Comparing the difference between 3 kg and 5 kg materials/batch, there is no significant change, and the efficiency is 1.7–1.71 mL/g, still highest at 130 °C. Therefore, the temperature chosen for that period is 130 °C.

The influence of hydrodistillation time

Extraction time is the third factor affecting the hydrodistillation of orange essential oil. The long enough time for water to diffuse the essential oil from the raw materials into the environment increased the yield of essential oils. In contrast, water exposed to heat for a long time generates heat and high pressure, which can degrade the yield of essential oil due to heat-sensitive compounds in essential oils, thermal decomposition, energy consumption, and cost manufacturing. Fig. 2(e-f) shows that the effect of time on the yield of essential oils tends to increase with increasing time from 30 to 60 min. This trend is true for both 3 kg and 5 kg scales. The highest yield of the 3 kg scale was 1.78 mL/g, while the 5 kg scale was 1.8 mL/g at 140 min. Therefore, to optimize the time factor of hydrodistillation of orange essential oil, 140 min is the extraction time selected to bring the highest efficiency.

Assessment of the Chemical Composition of Optimized OEO Samples

The factors affecting the hydrodistillation process of orange essential oil on equipment with a capacity of 50 L/batch after optimal investigation include: the size of the peels to be grounded, the materials and water ratio is 1:3 (g/g), hydrodistillation temperature of 130 °C, hydrodistillation time of 140 min with yields reaching 1.78 mL/g (3 kg scales) and 1.8 mL/g (5 kg scales). After running under optimal conditions to obtain essential oil, this essential oil was anhydrous with Na_2SO_4 and evaluated for chemical composition by the GC-MS method. Analytical results are presented in Table 1, Fig. 3, and Fig. 4. In this study, the essential oil of orange was analyzed and found to have 3 components, with limonene compound as the main component accounting for very high content of 98%, then β-Pinene (1.143 and 1.193%), and α-Pinene (0.720–0.785%). When comparing the analytical results between the 3 kg and 5 kg samples, there was no difference in the composition of the compounds, although there was no significant change in the content. This can also be explained by the fact that the essential oils are made from the same raw materials,

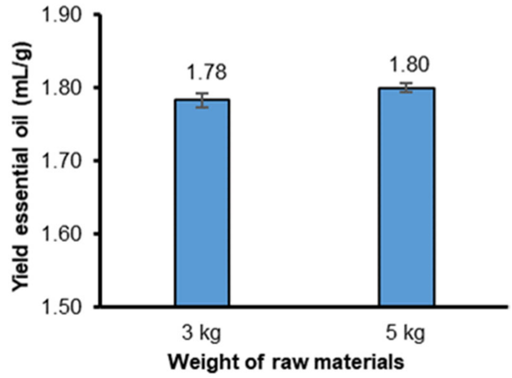


Fig 3. Comparison of the yield of essential oils obtained between 3 kg and 5 kg

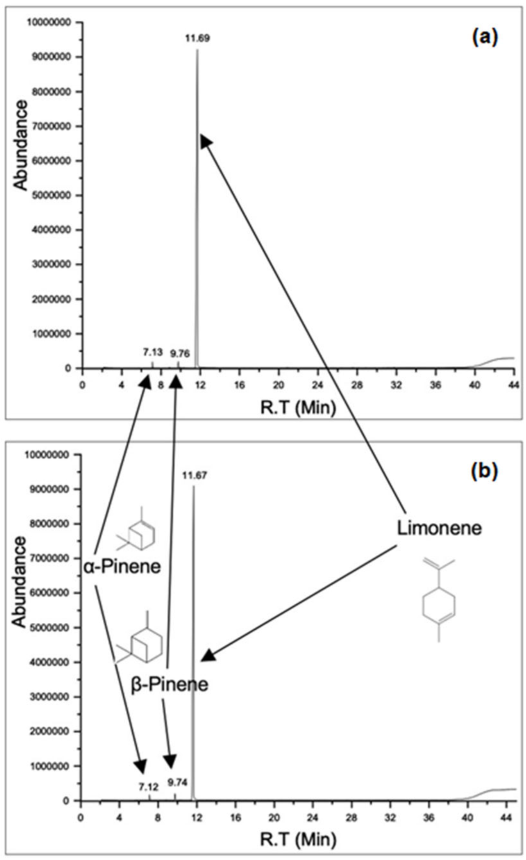
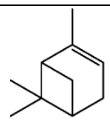
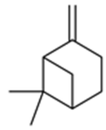
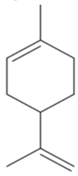


Fig 4. GC-MS spectrum of the composition of OEO (a) 3 kg scales (b) 5kg scales)

Table 1. Composition and content of compounds in orange essential oil

Peak	R.T (min)	Name compounds	Molecular formula	Structure formula	Content (%)	
					3 kg scales	5 kg scales
1	7.125	α-Pinene	$C_{10}H_{16}$		0.785	0.720
2	9.760	β-Pinene	$C_{10}H_{16}$		1.143	1.193
3	11.695	Limonene	$C_{10}H_{16}$		98.072	98.087

the same time of harvest, the same methods, conditions, and distillation equipment.

In addition, in Table 2–6, the content of volatile compounds in orange essential oil is compared in this study with essential oils of the Citrus family such as orange, lemon, grapefruit, and sweet mandarin. In these tables, only the content of compounds contained in this study is compared with other studies in many countries at different harvest times. In general, essential oils hydrodistillation from the fresh peels of Citrus trees often have limonene as the main compound, accounting for the highest content (over 70%). In addition, it can be seen that the Limonene compounds in Table 2–6 in different studies have the highest concentration in oranges (most of which account for more than 80% of the remaining compounds) compared with other types of citrus fruits.

Specifically, in Table 2, a comparison of the same species of *Citrus sinensis* shows that the Limonene compound accounts for a high content from 61.08 to 95.54%, followed by β-pinene accounted for from 0.25 to 5.45%, and α-pinene accounted for range from 0.3 to 5.98%. However, it is easy to see that the content of the main compound Limonene in this study is considered to be the highest compared to the cited studies. This is also understandable because the quality of the essential oil depends a lot on the changing factors such as the growing conditions of the orange trees, the harvesting conditions of the fruits, and the factors affecting the distillation method [24-25]. This was even more pronounced when the cultivars were changed by comparing the concentrations of compounds in the essential oils of orange (*Citrus sinensis*) and orange (*Citrus aurantium*). Results from Table 3 showed that limonene remained the main compound. However, the content is still lower than that of the essential oil distilled from orange peel (*Citrus sinensis*).

On the other hand, the study also compared the content of compounds in orange (*Citrus sinensis*) and lemon (*Citrus limon*) essential oils, with other lemon essential oils, with grapefruit (*Citrus paradisi*) and mandarin (*Citrus reticulate*), as shown in Table 4, 5 and 6. The results show that limonene is still the main compound, although there are many significant changes in some studies. That changes in the study of Kaskoos [1], Gomes et al. [41], Atti-Santos et al. [20], Smeriglio et al. [46], Sreepian et al. [47] when the content of the main compound Limonene accounted for less than 50% in the essential oils of lemons. This is the opposite in the essential oils of *Citrus paradisi* peels and *Citrus reticulate* peels, the main compounds in these peels all account for high content of over 80%.

Therefore, the main compound present in orange essential oil in Vietnam in this study is limonene, accounting for more than 98%, higher than in previous studies. This also helps other studies to show that oranges contain high amount of limonene, which has good fragrance, antibacterial and antioxidant properties in food, cosmetics, and pharmaceutical products [26-27].

Table 2. Comparison of compounds in orange essential oil (*Citrus sinensis*)

		Citrus sinensis											
	In this study	2006 [28]	2009 [29]	2012 [30]	2017 [17]	2017 [31]	2019 [32]	2020 [15]	2020 [14]	2020 [33]	2020 [34]	2020 [35]	2021 [36]
	Vietnam	Pakistan	India	Tunisia	Nigeria	Turkey	China	Iran	South Africa	Algeria	Turkey	Egypt	China
α-Pinene	0.785	0.84–1.26	0.291–0.390	0.07-0.44	0.7	0.9	5.98	1.85	0.9	0.53	0.32	0.3–1.4	0.33
β-Pinene	1.143	5.45	3.043–3.443	0.97–1.80	2.7	0.05	-	-	-	0.25	-	-	1.9
Limonene	98.072	61.08 -76.2	35.060–76.68	81.52–86.43	92.14	92.01–93.32	90.75	71.54	80.5	95.54	88.90	74.5–82.1	88.25

Table 3. Comparison of compounds in the essential oils of orange (*Citrus sinensis)* and orange (*Citrus aurantium*)

	Citrus aurantium					
	In this study	2016 [37]	2018 [18]	2019 [5]	2019 [38]	2021 [10]
	Vietnam	Iran	Croatia	Bulgaria	Iran	Turkey
α-Pinene	0.785	0.30	0.8	1.29	1.32	-
β-Pinene	1.143	0.65	0.8	0.07	0.97	0.26–0.28
Limonene	98.072	94.81	91.1	85.22	85.49	72.51–95.70

Table 4. Comparison of compounds in orange (*Citrus sinensis*) and lemon (*Citrus limon*) essential oils

	Citrus limon								
	In this study	2006 [27]	2010 [39]	2012 [30]	2019 [1]	2019 [3]	2020 [40]	2020 [41]	2021 [42]
	Vietnam	Pakistan	Korea	Tunisia	Iraq	Sardegna	India	Brazil	Bulgaria
α-Pinene	0.785	2.63	2.6	0.13–5.9	2.25	0.43	00.17	2.4	-
β-Pinene	1.143	10.80	15.7	0.63–31.49	23.89	-	-	23.01	1.5
Limonene	98.072	53.61	64.54	37.63–69.71	29.52	77.44	55.40	44.75	82.96

Table 5. Comparison of compounds in orange essential oil (*Citrus sinensis*) and other lemon essential oils

	In this study	*Citrus meyerii* 2005 [43]	*Citrus latifolia* 2005 [20]	*Citrus karna* 2009 [29]	*Citrus limettioides* 2012 [44]	*Citrus bergamia* 2012 [45]	*Citrus lumia* 2018 [46]	*Citrus hystrix* 2019 [47]	*Citrus aurantifolia* 2020 [22]
	Vietnam	Italy	Brazil	India	India	Tunisia	Italy	Thailand	Vietnam
α-Pinene	0.785	1.33–2.25	1.9	1.227	0.865	0.48	0.62	2.22	1.855
β-Pinene	1.143	1.04–16.90	12.4	1.8	2.933	4.38	6.89	21.10	11.287
Limonene	98.072	51.95–79.7	47.5	92.310	89.089	59.21	48.91	25.28	65.988

Table 6. Comparison of compounds in the essential oils of orange (*Citrus sinensis*), *Citrus paradise,* and *Citrus reticulate*

	In this study	*Citrus paradise*							*Citrus reticulate*		
		2006 [27]	2010 [39]	2015 [9]	2020 [48]	2020 [21]	2021 [42]	2021 [49]	2012 [30]	2021 [42]	2021 [36]
	Vietnam	Pakistan	Korea	Argentina	China	Algeria	Bulgaria	Algeria	Tunisia	Bulgaria	China
α-Pinene	0.785	1.26	1.0	0.60	0.76	0.89	-	1.08	0.39–1.25	-	0.62–1.22
β-Pinene	1.143		-	-	0.05	1.06	1.13	-	0.06–1.59	1.01	2.00
Limonene	98.072	86.27	94.20	92.60	93.33	87.51	91.78	67.22	51.81–69.0	84.88	79.13–91.54

In addition, this can also be shown that because each geographical region, climatic conditions, farming, and harvesting conditions will greatly affect the quality as well as the content of volatile compounds present in the plants.

Assessment of Performance of Hydrodistillation Equipment 50 L/Batch

In order to evaluate the stable operation of the oil hydrodistillation equipment with the capacity of 50 L/batch, this study carried out 3 times of 5 kg scales with optimal conditions. The yield of the obtained essential oil was 1.8–1.81 mL/g. The results shown in Fig. 5 and Table 7 show that after 3 times of hydrodistillation at 5 kg, the yield of the obtained essential oil did not change significantly (almost 1.8 mL/g) due to errors in the extraction process, making the essential oil anhydrous. In terms of color and scent, the essential oils obtained through 3 times of applications have the same color and characteristic scent of orange. When analyzing the content of compounds in orange essential oil by GC-MS method, 3 components were detected, with Limonene compound being the main component accounting for a high content of 98.144–98.182%, α-pinene (0.655–0.734%) and β-pinene (1.114 and 1.163%). Most of these compounds changed insignificantly over the measurements. This shows that this 50 L capacity essential oil distiller works stably and obtains good quality and content without changing the yield and volatile compounds content.

Through the process of orange essential oil hydrodistillation on this equipment 50 L, it was found that the equipment is easy to operate, stable, does not cause noise, and recovers the yield of the essential oil between scales. However, the device still has some shortcomings, such as the small input material space, which makes it difficult to input peels, and the small place where the residue is left after distillation, so the process of getting the residue is still hindered.

■ CONCLUSION

This study investigated the factors affecting the hydrodistillation of orange essential oil on a 50 L/batch device with a weight scale of 3 kg and 5 kg. The results show that after pre-treatment, the orange peels are ground into small pieces, put into the raw material tank of the distillation device, the amount of water is added to the distillation system at a ratio of 1:3 (g/g) at a heating temperature of 130 °C and a distillation time of 140 min. A comparison of the content of the essential oil compounds obtained on the 3 kg and 5 kg scales is also provided. The results show that at the above optimal conditions, the yield of the obtained essential oil did not change significantly (about 1.8 mL/g), and the volatile components in the essential oils of these two scales also did not change in composition and content. Accordingly, Limonene compounds are the main compounds accounting for over 98%. Therefore, the device is rated as easy to operate, stable, does not cause noise, and recovers a stable yield of essential oil between implementation times.

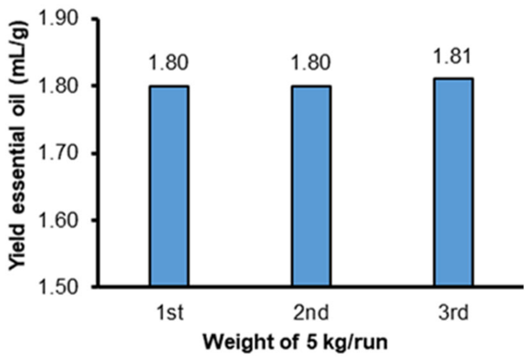


Fig 5. The yield of essential oil obtained after 3 times by 5 kg scales

Table 7. Composition and content of compounds in orange essential oil at 5 kg scales

Peak	R.T (min)	Compounds	Content (%)		
			1	2	3
1	7.136	α-Pinene	0.717	0.655	0.734
2	9.781	β-Pinene	1.114	1.163	1.123
3	11.705	Limonene	98.170	98.182	98.144

■ ACKNOWLEDGMENTS

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