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RESEARCH PAPER

Hydrodistillation of essential oil from peels of orange (*Citrus sinensis*) in the Mekong Delta, Vietnam: process optimization and chemical profiling

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Abstract

In this study, essential oils were extracted from peels of orange (*Citrus sinensis*) by employing hydrodistillation. The materials were collected in Ben Tre Province, Vietnam and the process was optimized by varying some experimental parameters including temperature, water-to-material ratio, extraction time and material size. The obtained essential oil was analyzed by Gas Chromatography-Mass Spectrometry to determine chemical composition. The maximal essential oil yield was determined as approximately 4%, obtained at the following conditions: ratio of water and lemon peels of 3:1 (mL/g), 60 mins and130°C. GC-MS results revealed that Limonene (98.454%), β-Myrcene (0.997%), and α-Pinene (0.549%) are the major constituents in the essential oil.

1. Introduction

Nowadays, the extraction of natural compounds in medicinal herbs is one of the topics attracting interest from botanists but also scientists (Kim Ngan, Huong, Le *et al*., 2019; Kim Ngan, Thu Thuy, Tuyen *et al*., 2019; Hien *et al*., 2019; Ngan *et al*., 2020). In particular, plants containing essential oils of the Citrus family are one of the families with many trees of high economic value, with many applications used as spices, raw materials in the industry of fragrance, cosmetics, and pharmaceuticals (Hien *et al*., 2018; Kim Ngan *et al*., 2019; Quyen *et al*., 2020; Ngan *et al*., 2021). Many bioactive compounds in essential oils have been detected such as aldehydes, alcohols and esters, and other aromatic compounds. Citrus is naturally distributed in India, southern China, Vietnam, Laos, Cambodia, Thailand, and Myanmar, in which the richest and most diverse centres are India and Malaysia (Mabberley, 2004). In Vietnam, the genus Citrus has about 20 species and many varieties are grown in most regions (Ngan *et al*., 2021). Citrus varieties are grown mainly for their fruit, in addition, the peels, flowers, and leaves of many varieties are used to extract essential oils and separate flavonoid compounds with high biological activity such as antioxidant, anti-inflammatory, antibacterial, inhibiting cancer cells and preventing skin diseases (Kademi and Garba, 2017; Toscano-Garibay *et al*., 2017; González-Mas *et al*., 2019; Singh *et al*., 2020). Orange scientific name: *Citrus sinensis* is a species of flowering plant in the family Rutaceae. Small tree, thornless or few thorns. Leaves alternate, longleaf blade, dark green, oval. Inflorescences short, axillary, solitary, or in groups of 2-6 flowers in clusters, fruit nearly globose. Orange essential oil is a clear, pale yellow liquid with a very pleasant characteristic aroma. In essential oils, the main component of essential oil is D - Limomen accounting for over 85%, besides there are some other substances such as alpha-pinene, Beta-pinene, sabinene, limonene, b -ocimene, linalool (Allaf *et al*., 2013; Chen *et al*., 2014; O'Bryan *et al*., 2008).

In this study, the essential oil from orange peel was recovered by hydrogen distillation or steam distillation, with an easy-to-implement and simple experimental procedure and reduced loss of essential oil during the extraction process, high efficiency (Samadi *et al*., 2017; Hall *et al*., 2018). The key factors in the identification and optimization are extraction time, energy consumption, quality, and yield of the essential oil. The

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choice of the best method depends on a compromise between different criteria. Therefore, this study mainly focuses on investigating the conditions affecting the extraction process of essential oils such as extraction time, water-to-material ratio, and temperature by a single -factor experiment. Furthermore, we also determined volatile components of essential oil from orange (*Citrus sinensis*) leaves grown in Ben Tre province, Vietnam by GC-MS. The research results of the project will complement the research results on the chemical composition of orange essential oil and reinforce a new research direction on the chemical composition of citrus species, contributing to the discovery of other compounds the natural substance with high medicinal properties.

2. Materials and methods

2.1 Raw materials and essential oil extraction

Fresh *Citrus sinensis* (orange) was harvested in the Mekong Delta, Vietnam. The oranges were pre-treated with water and peeled. The pulp and the shell are kept separate. The peel is used to extract the essential oil, 50 g of orange peel is pureed into a container of 1 L of distilled water. The Clevenger-type hydrodistillation system is operated for 30-240 mins. At the end of the process, the crude oil is anhydrous to remove water with Na_2SO_4 and stored under 4ºC conditions. The yield of orange oil obtained (%) was calculated according to the following formula:

Yield of essential oil (%) = (Mass of essential oil obtained (mL))/(amount of raw materials used (g)) × 100%

2.2 Effect of extraction conditions

To determine the optimal extraction conditions of *Citrus sinensis* (orange) peel essential oil, the conditions of ratio, temperature, and extraction time were investigated and evaluated by distillation with water solvent. Specifically, the experimental process was carried out based on the evaluation of the influencing parameters of the ratio of water to raw materials (2:1, 3:1, 4:1, 5:1, and 6:1 mL/g), extraction time (15, 30, 45, 60, 75 and 90 mins) and change in extraction temperature (100, 110, 120, 130 and 140°C).

2.3 Analysis of chemical composition GC-MS

Volatile compounds in essential oil samples were analyzed using an Agilent 6890 N GC instrument (Agilent Technologies, Santa Clara, CA, USA) combined with an inert HP5-MS and MS 5973 column. The column head pressure is 9.3 psi. 25 µL of essential oil was added to 1.0 mL of n-hexane and dehydrated with Na_2SO_4. The flow rate is constant at 1 mL/min. The nozzle temperature was 250°C and the fractionation rate was 30. The area percentage and retention time of the components were determined using the Wiley 275 L data library of the GC-MS system.

3. Results and discussion

3.1 Optimizing the extraction performance of orange essential oil

3.1.1 Effect of material size

The processing of the raw materials greatly affects the yield and quality of the essential oil when producing the finished product (Figure 1). The experiment was conducted on three basic sizes of raw materials (origin, cut fibre and grind), with significant changes in the yield of essential oils recovered with values of 1.6%, 1.8% and 2.4%, respectively. It can be seen that the processing of raw materials with fine grinding facilitates the extraction of essential oils during the distillation process. At the same time, reducing the useful capacity of the device and steam escaping through the mass too quickly will reduce the efficiency of essential oil separation. The most suitable grinding of raw materials is through the sieve of about 3-5 mm, thereby shortening the distillation process because the essential oil is released more easily when exposed to steam, so it will improve the efficiency of the extraction process. essential oil extraction. Therefore, the grinding method used for material processing is the most suitable.

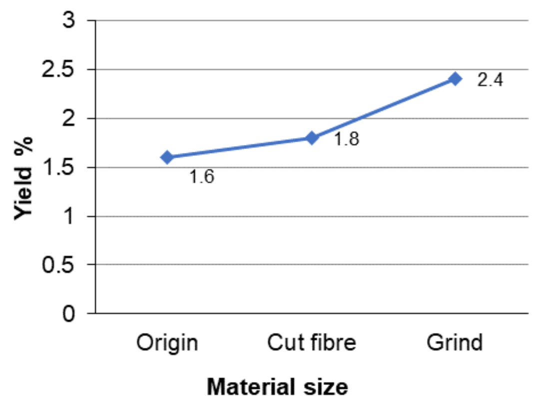

Figure 1. Effect of material size on essential oil yield of orange (*Citrus sinensis*) peel

3.1.2 Effect of extraction temperature

Temperature is one of the factors that significantly affect the quality of essential oils. Figure 2 shows that the efficiency of essential oil recovery depends on the increase in temperature. Specifically, when gradually increasing the temperature from 100°C to 140°C, the yield of essential oil also increased from 2% to 2.9%. However, at a temperature of 130°C, the efficiency was 2.9% and there was no change when increasing the temperature to 140°C. When the temperature is

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increased, the osmotic diffusion will increase, and the solubility of essential oils in water also increases, but the decomposition of essential oils also increases because most essential oils are less stable under the effect of heat. Therefore, the distillation temperature at 130ºC was selected for further experiments.

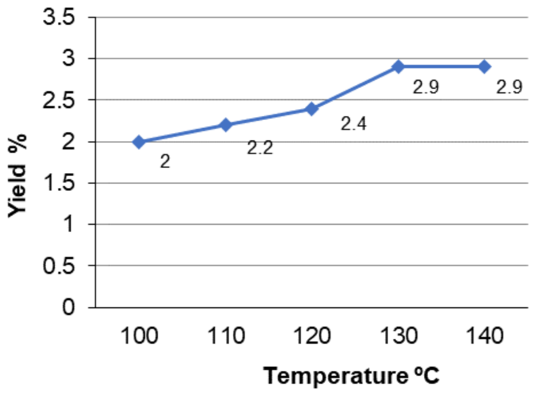

Figure 2. Effect of extraction temperature on essential oil yield of orange (*Citrus sinensis*) peel

3.1.3 Effect of raw material and solvent ratio

During the extraction of essential oils, when heating the mixture of raw materials and water, steam penetrates the cell layers, breaking the essential oil bag and attracting the essential oil to the steam. This process is repeated until the essential oils in the tissues are completely drained out. Thus, the presence of water is very necessary, if the amount of water is too small, it will not dissolve the glue and salt surrounding the essential oil bag, making the essential oil unable to escape. But if the amount of water used is too much, it is not beneficial, especially in the case of essential oils that contain water-soluble components. In this study, the change in the ratio of water and raw materials was shown by the yield of the recovered essential oil in Figure 3. Typically, when increasing the solvent/material ratio from 2:1 to 3:1, the efficiency increases from 3.2% to 3.4%. When continuing to increase the ratio of solvent/ raw materials, the efficiency of essential oils showed a clear sign of decline. The results show that, at a certain limit, the amount of essential oil recovered will not

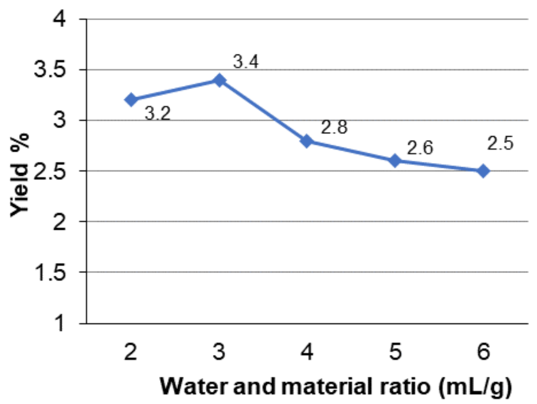

Figure 3. Effect of water and material ratio on essential oil yield of orange (*Citrus sinensis*) peel

increase significantly despite increasing the amount of solvent. If the excess solvent is used, it will cause waste, waste solvent, increase the amount of impurities, and waste energy for the solvent recovery process. Therefore, the 3:1 solvent/raw material ratio is suitable to extract the maximum amount of essential oil in the material.

3.1.4 Effect of extraction time

One of the factors that directly affect the quality of orange essential oil is the extraction time. Based on the results obtained in Figure 4, it was shown that up to a certain time threshold, increasing extraction time did not increase the extraction efficiency of essential oils. Specifically, when increasing the extraction time from 15 mins to 60 mins, the yield increased continuously and reached the highest yield of 4%, continuing to increase the time to 90 mins, the yield of essential oil decreased to 3.7%. Although, the longer the distillation time, the more thoroughly the essential oil is obtained, prolonging the time may not increase the amount of essential oil, but can affect the quality of the essential oil and be costly in terms of time and energy. Therefore, 60 mins are suitable for the optimal extraction process of orange essential oil.

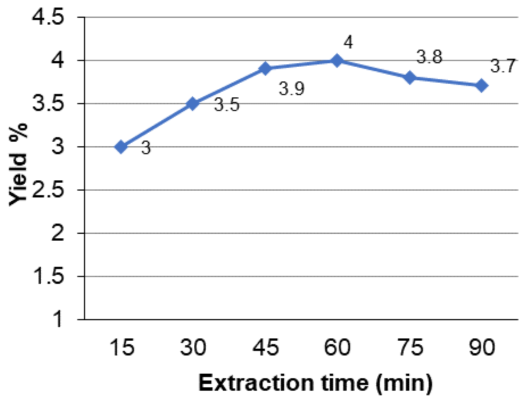

Figure 4. Effect of extraction time on essential oil yield of orange (*Citrus sinensis*) peel

The study results demonstrated that different conditions directly affect and play an important role in optimizing the experimental conditions of the hydrodistillation extraction method. The main parameters affecting the yield of essential oils are the size of the material, the temperature, the ratio of water to the material, and the extraction time. The highest extraction yield was obtained in the test with crushed material, ratio 3:1, heating at 130°C for 60 mins. When the different conditions were changed, the extraction yield was significantly different. The highest yield of distillation is 4% under optimal conditions.

This result is consistent with previous research results, in which the yield of essential oil extracted from orange peel ranges from 0.42-4%. In Valencia, Mohamed (2006) conducted extraction studies by hydrodistillation and microwave-assisted hydro-

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distillation, with essential oil yields of 0.42 and 0.39%, respectively. Limonene is the most abundant component in monoterpene hydrocarbons 76.7% (HD) and 78.5% (MAHD), along with some β-myrcene, α-sisensal, linalool and decanal components (Ferhat *et al.*, 2006). In Iraq, Ibtehal *et al.* (2015) conducted a survey and evaluation of citrus essential oils based on two methods, microwave-assisted steam distillation (MASD) and steam distillation (SD). In which, an orange essential oil with SD and MASD distillation methods, the yield was 1.095% and 1.091%, respectively. Besides, the research team has shown the difference in limonene composition between SD (83,2189%) and MASD (80,9661%) by the unique characteristics of each extraction method (Shakir *et al.*, 2015). Hydro-distillation is carried out under optimal conditions to achieve good performance. Various factors are affecting the yield of essential oils including harvest time, growing stage, different parts, and environmental conditions (Turek and Stintzing, 2013; Naeem *et al.*, 2018). That is the reason for the different yields of essential oils when extracting the same method.

3.2 Essential oil ingredients

To more accurately assess the quality of the obtained essential oil, analyze and determine the volatile compounds in the essential oil by GC-MS method. The results shown in Table 1 and Figure 5 shows the chemical composition and percentage of orange essential oil, the main compounds identified as Limonene (98.454%), β-Myrcene (0.997%), and α- Pinene (0.549%).

Figure 5. Chromatography of orange peels essential oil from the hydrodistillation method.

Peak	R.T. min	Compounds	This study
1	7.387	α-Pinene	0.549
2	10.095	β-Myrcene	0.997
3	12.103	Limonene	98.454

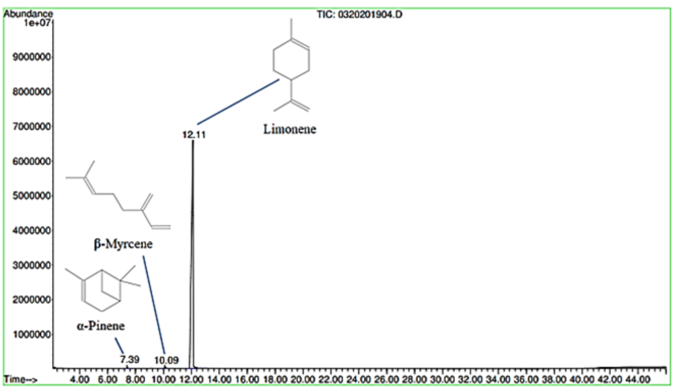

Figure 5. Chromatography of orange peels essential oil from the hydrodistillation method.

The quality of essential oils can be negatively affected by the use of pesticides and other chemicals, changes in altitude, soil conditions, and rainfall. Each type of tree must be grown in the appropriate climatic and environmental conditions for that type of tree to obtain the best quality raw materials. Harvesting essential oil plants in the early morning or late afternoon is also quite important in ensuring the quality of the essential oil. For example, Velázquez-Nuñez *et al.* (2013) conducted a study on the chemical compounds and anti-fungal effects of *Aspergillus flavus* of essential oils obtained from oranges in Puebla (Mexico) in which limonene is the main component accounting for 96.62% of the essential oil, along with some components identified as α-pinene, β-pinene, β-myrcene, and citral Z and E (Velázquez-Nuñez *et al.*, 2013). In the study elsewhere, oranges grown in Kenya were reported by Njoroge *et al.* (2005) isolation by cold pressing and evaluation of volatile components in essential oils. The results showed that monoterpene hydrocarbon compounds dominated mainly in the range of 92.7% - 96.9% and 0.1% of sesquiterpene hydrocarbons. Limonene, sabinene, α-terpinene, and α-pinene are the main compounds (Njoroge *et al.*, 2005).

In 2011 publication by Farhat *et al.* also showed a difference in the extraction process of essential oils when using two methods of microwave steam diffusion (MSDf) and steam diffusion (SDf). The yield of essential oil obtained by the two methods MSDf and SDf was 1.54% and 1.51%, respectively. Similar to other publications, the dominant compounds in orange essential oil from Spain are still Monoterpenes with the main component being Limonene (MSDf 94.88% and SDf 95.03%) (Farhat *et al.,* 2011). In addition, Tao *et al.* (2009) used materials from oranges in Xiangtan (China) to recover essential oil based on the steam distillation method. The research team used the obtained essential oils to evaluate the antibacterial activity against several strains of bacteria such as *Staphylococcus aureus, Escherichia coli*, and *Bacillus subtilis*. Based on the presence of major components typical in the essential oil of Limonene, followed by sabinene, myrcene, g-terpinene, and α-pinene (Tao *et al.*, 2009). Limonene (the main compound) plays an essential role in protecting plants from insect attacks. Furthermore, Limonene can also be used as a fragrance ingredient for cosmetic products (Sun *et al.*, 2007). Previous work showed that β -Myrcene is a naturally occurring base chemical in stable chemistry. Furthermore, Myrcene is an important intermediate applied in the perfume industry (Behr *et al.*, 2009). In addition, α-Pinene is an antibacterial and anti-inflammatory agent (Silva *et al.*, 2012). In conclusion, the essential oil of orange peel has great potential for application as a natural insecticide based on its identified compound.

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4. Conclusion

Through the process of surveying and optimizing oil extraction conditions, the highest yield of essential oils was 4% (v/w). Using single-factor investigation, the essential oil from *Citrus sinensis* (orange) peel was extracted based on a successful optimized hydrodistillation method with 100 g of raw materials, the ratio of water and raw materials was 3:1. mL/g was heated at 130°C for 60 mins. GC-MS analysis of the chemical composition of essential oil from orange peel (*Citrus sinensis*) grown in Ben Tre province, Vietnam showed that limonene, β-myrcene, and α-pinene content in orange essential oil reached 98.545%, 0.997%. and 0.549%, respectively. Therefore, the results of this study contribute to the orientation for building a pilot scale-up process in extracting essential oils of the citrus family, thereby applying to improve the value and economic efficiency of local orange trees direction.

Conflict of interest

The authors declare no conflict of interest.

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