Journal of
Patchouli and Essential Oil Products

JPEOP

http://doi.org/10.24815/jpeop.v1i2.25408 Vol. 1, No. 2, Page 36-41, December 2022

Vacuum Distillation of Aceh Patchouli Oil into Hi-Grade and Crystal Patchouli with Rotary Vacuum Evaporator

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Received: 24 Maret 2022; Accepted: 15 December 2022; Published online: 24 December 2022

Abstract

Patchouli or *Pogostemon cablin* benth is one of the most important essential oil-producing plants in Indonesia. In the world of trade, patchouli oil is widely used as raw material, a mixing agent, and a fixative. Patchouli oil can be upgraded to Hi-Grade Patchouli (HGP) with a Patchouli alcohol (PA) content of 40-80% through vacuum distillation technology. Furthermore, HGP can be further processed into crystal patchouli with a purity of up to 90-100%. This study aims to improve the quality of patchouli oil produced by the community. This study examines the effect of variations in vacuum pressure on the distillation process on the quality of the resulting patchouli oil, determines the impact of cooling temperature variations on the resulting crystal mass, and determines the crystal quality (patchouli alcohol content and crystal size). The method used was vacuum redistillation followed by crystallization of patchouli oil. In the redistillation stage, it is carried out using a rotary vacuum evaporator. Redistillation is conducted in two stages: the first is to produce a light fraction at a temperature of 125 °C, and the second is to produce a heavy fraction at a temperature of 140 °C. The variable that was varied was process pressure. The pressure used is <1; 2-3; 6-8 and 10-12 mbar. The process is continued with crystallization using the cooling method (cooling crystallization) with a cooling temperature variation of -12 °C and -15 °C. The results showed that through the vacuum redistillation method, the quality of patchouli oil produced by the community could be increased in PA levels up to 81.79%. The lower the operating pressure, the highest the quality of patchouli oil is produced. The lower the crystallization cooling temperature, the more crystal mass is formed. The crystal purity of this HGP resulted in very high levels of crystalline PA, namely 99.6%. The crystal size obtained is 290-600 µm on average.

Keywords: essential oils, patchouli oil, vacuum redistillation, rotary vacuum evaporator, cooling crystallization

1. INTRODUCTION

Essential oil is one of the agricultural products which has great prospects in the future. Nowadays, there are 70 types of essential oil that have been traded on the international market, and Indonesia has 40 kinds of plant that produces essential oil. However, only 14 types have a real role as export commodities (Hetik et al. 2013). Patchouli or *Pogostemon cablin* benth is one of the most important essential oil-producing plants in Indonesia. Patchouli oil is widely used as raw material, mixing, and fixative material (fragrance binder) in the perfume, pharmaceutical, and cosmetic industries (Maisarah et al. 2017; Isnaini et al. 2022).

One of Aceh's patchouli industry problems is that the local community receives less profit because all patchouli oil is exported in the form of crude patchouli oil. Currently, patchouli farmers in Aceh only use simple steam distillation equipment to obtain patchouli oil, while enhancing the quality of patchouli oil requires a follow-up step. One of the advanced processes for refining patchouli oil can be done by redistilling (re-distillation of patchouli oil) by evaporating the distilled patchouli oil (Nurjanah et al. 2016).

According to Sufriadi and Mustanir (2004), to get high-quality patchouli oil, it must have a high amount of patchouli alcohol. But in fact, patchouli oil produced by the local farmers still has lower levels of patchouli alcohol which is below 30%. Based on this condition, patchouli oil produced by the local farmer does not fulfill export quality standards. Through vacuum distillation technology, patchouli oil can be upgraded to Hi-Grade Patchouli (HGP) with 40-80% of Patchouli alcohol (PA) levels (Adani and Pujiastuti 2017).

In various chemical industries, such as the perfume industry, requires a high quality of patchouli oil (van beek and Joulain 2018). Patchouli oil produced from purification using vacuum distillation can produce patchouli alcohol above 70%. Then continued with the process of crystallization of patchouli oil which also plays an essential role in the chemical industry because 70% on average of chemical products produced by industry are solids or crystals. The advantages of crystal or solid-shaped products are cheaper at transportation costs, not easily damaged due to decomposition, and easier to pack and store (Setyopratomo et al. 2003).

This research aims to improve the quality of patchouli oil produced by local community. Additionally, this research examines the effect of vacuum pressure variations on the quality of patchouli oil produced to determine the impact of cooling temperature variation on the mass of crystals and determine the quality of the crystal (patchouli alcohol levels and crystal size).

2. MATERIALS AND METHODS

2.1 Material

This research was conducted at the laboratory of Atsiri Research Centre (ARC) and Laboratory of Resource and Energy, Department of Chemical Engineering, Faculty of Engineering, Universitas Syiah Kuala. The research methods used are vacuum redistillation and cooling crystallization. Figure 1 shows the research scheme. The raw material used in this research is patchouli oil originating from Aceh Jaya with 36% of initial PA rate, and the chemical material used is KOH. The tools used in distillation process are rotary vacuum evaporators, vacuum pumps, and scales. While in the crystallization process is freezer, infrared thermometer, chemical glass, and measuring glass. Then in the sample testing process using GC-MS, SEM, buret, picnometer and refractometer.

2.2 Vacuum Redistillation Process

Patchouli oil from farmers is carried out with vacuum redistillation process to get patchouli oil with higher levels of Patchouli Alcohol. In this process using a Rotary vacuum evaporator with the first distillation temperature of 125 °C, the second distillation temperature of 140 °C, a rotation speed of 80 rpm, 300 grams of patchouli oil mass, and a vacuum pump with a variation in pressure of <1; 2-3; 6-8; 10-12 mbar. The first redistillation produces light fractional patchouli oil, and the second redistillation produces heavy fractional patchouli oil. Light fractional patchouli oil has smaller levels of patchouli alcohol, and heavy fractional patchouli oil produces the greatest levels of patchouli alcohol. Heavy fractional patchouli oil was used as a sample that will be crystallized.

2.3 Cooling Crystallization Process

Cooling crystallization is performed using 20 grams of distillate mass at temperatures ranging between -12 and -15 °C and stirring speeds of 50 rpm. Before the crystallization process, Hi-grade patchouli oil was weighed first. The oil is weighed and put in a 100 ml chemical glass. The oil is placed in the freezer and stirred after a period of rest. After a few days, physical observations are conducted, specifically the saturation of the sample to the level when crystal clusters form. It is then transferred into a filter paper with a buffer (sieve) for the separation process and left at room temperature for 24 hours. The result of separation produces white crystals with high purity.

2.4 Sample Testing Process

Hi-Grade patchouli oil is analyzed using Gas Chromatography Mass Spectrometry (GC-MS) to determine the levels of patchouli alcohol and other analyses to determine the refractive index, type, weight, and acid rate. The crystals were analyzed using several analytical tools, namely: Gas Chromatography Mass Spectrometry (GC-MS) to find out the level of 7 patchouli alcohol, and Scanning Electron Microscope (SEM) to find out the size of the crystal.

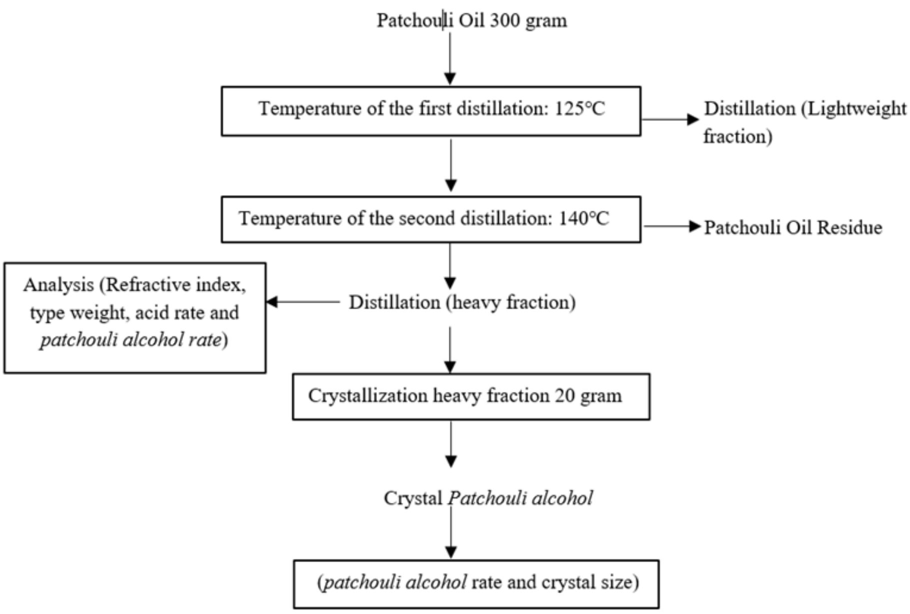


Figure 1. Research scheme

Table 1. Distillate results in light and heavy fraction vacuum distillation experiments with fixed variables

Pressure (mbar)	Temperature (°C)	Refractive Index	Type Weight (g/mL)	Acid Number	Patchouli Alcohol (%)
<1	125	1.506	0.93	2.083	-
	140	1.5115	0.982	5.971	81.79
2-3	125	1.504	0.924	1.805	-
	140	1.511	0.974	4.305	74.23
6-8	125	1.5025	0.916	1.527	-
	140	1.510	0.964	3.333	68.59
10-12	125	1.502	0.91	1.389	-
	140	1.508	0.948	2.777	51.91

3. RESULTS AND DISCUSSIONS

Based on this research, we collected some data about the quality of patchouli oil after redistillation process. The used indicators are refractive index, type weight, acid number, and patchouli alcohol levels. Data on the quality of redistillation of patchouli oil can be seen in Table 1.

3.1 Quality of Patchouli Oil after Redistillation Process

3.1.1 Refractive Index

This indicator shows that the quality of patchouli oil by its purity levels. If patchouli oil is mixed with water; as a result, the refractive index becomes low. The refractive index number of the redistillation results is affected by the pressure of the process. This result can be seen in Figure 2 as below.

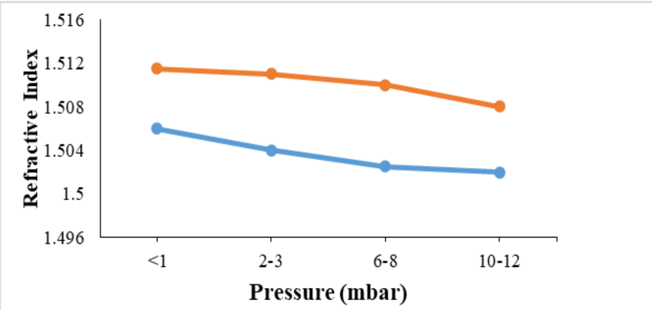


Figure 2. Effect of refractive index of patchouli oil redistilled results on light (blue line) and weight fractions (orange line)

The Patchouli oil sample on light fraction and weight fraction showed us it would decrease the refractive index value as long as the pressure increases. According to Raoult's law, the amount of distillate produced can be calculated by comparing the partial pressure of the desired compound with the total pressure of the mixture. The lower the pressure, the more fractions of the heavy component that come out as distillate (light fraction). It makes fewer light compounds left behind as bottom products. This condition causes the refractive index to be higher in the light and heavy fractions with lower pressure.

The essential oil refractive index is related to the components arranged in the essential oil obtained. The more long-chain such as sesquiterpenes or oxygen-eroding components that come out as distillate products, the density of essential oils will increase so that the value of the oil refractive index becomes higher. The refractive index values in the light fraction are in the range of 1.502–1.506 and 1.508-1.5115 in the weight fraction.

3.1.2 Density

Density also shows the quality of patchouli oil and is often associated with the components contained in it. The heavier components contained in the oil, the higher the value of the type of weight. In type weights, the oxygenated terpene component is higher than un-oxygenated terpenes (Idris et al. 2014). The weight type from the patchouli oil that has been redistilled can be seen in Figure 3.

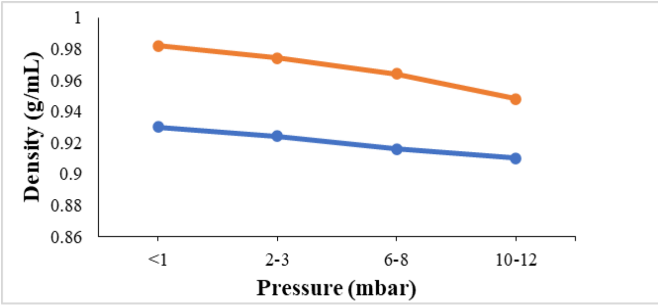


Figure 3. Effect of density of patchouli oil redistilled results on light (blue line) and weight fraction (orange line)

Figure 3 shows that samples of light and weight fractions of patchouli oil have decreased their weight values of type as the pressure increases. Similar to the refractive index, the components contained in patchouli oil also affect the value of type weights.

3.1.3 Acid Number

The number of acids indicates the level of free fatty acids (FFA) in patchouli oil. The increasing number of acids can affect the quality of patchouli oil, as acid compounds can change the original smell of patchouli oil (Idris et al. 2014). The acid number of the redistilled patchouli oil can be seen in Figure 4.

The chart above explains that the acid number will drop with the pressure increase and the decrease in the weight fraction more significant. The trend for this case is different from the case of refractive index and weight type, as discussed earlier. In the previous case, it was seen that the composition of the two fractions was linear. In this case, non-linear phenomena are more dominant. A decrease in the number of acids at the lowest pressure will result in more free fatty acids. Free fatty acids produced through the oxidation process of patchouli oil components, especially from the aldehyde group, can form carboxylic acid groups.

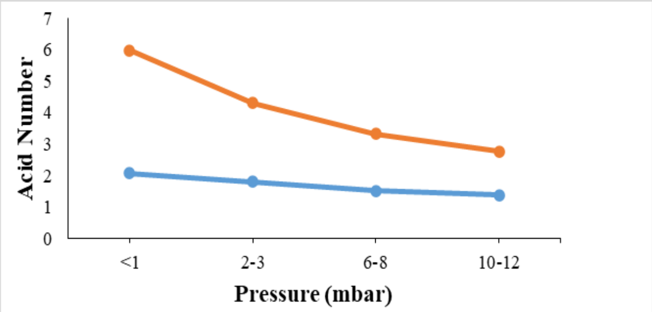


Figure 4. Effect of acid number of patchouli oil redistilled results on light (blue line) and weight fraction (orange line)

This process tends to occur at low pressure. The decrease in the number of acids is not very significant in light fractions because it contains a small aldehyde. The acid number of this study in the light fraction was in intervals of 1.389-2.083 mgKOH/g. For the weight fraction, the number of acids obtained is in the range of 2.777 – 5.971 mgKOH/g.

3.1.4 Acid Number

Patchouli alcohol levels are one of the parameters that determine the quality of patchouli oil (Kusuma et al. 2018). The higher the level of patchouli alcohol, the better the quality of patchouli oil. In this study, only the content of patchouli alcohol in the weight fraction was analysed. This is because the material of the weight fraction becomes the object in the next process. Patchouli alcohol levels from redistillation process can be seen in Figure 5.

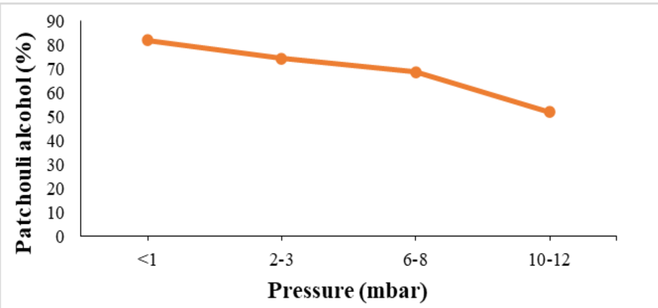


Figure 5. Patchouli alcohol levels in the heavy fraction of patchouli oil resulting from redistillation.

From Figure 5, it can be seen that the level of patchouli alcohol decreases as the process pressure increases. For each process pressure, PA levels are known as follows: 81.79; 74.23; 68.59, and 51.91%. The light component evaporates rapidly at low pressure. While at high pressure, the slightest light component evaporates. A lower percentage of PA at high pressure indicates more light components contained in the weight fraction. At a pressure of 10-12 mbar of distilled oil are mostly components whose boiling point is below the boiling point of patchouli alcohol such as terpene compounds (beta-patchoulen, anisole, and transcaryophyllen), so that the PA level in patchouli oil is 10-12 mbar less distilled.

3.2 Patchouli Alcohol Crystal

3.2.1 Effect of Cooling Temperature on The Mass of Crystals Obtained

The formation of PA crystals is carried out using a freezer on distillate patchouli oil as much as 20 grams at a PA level of 81.79; 74.23 and 68.59% with two variations in temperature, namely -12 °C and -15 °C, then cooled for three days. The relationship between the RATE of PA and the mass of the crystal obtained can be seen in Figure 6.

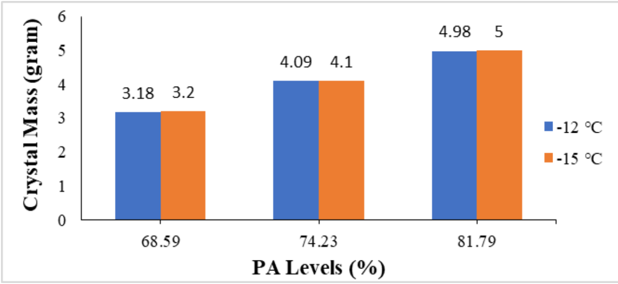


Figure 6. Effect of cooling temperature on the mass of crystals obtained

From the figure above, it can be seen that the mass of crystals obtained increases along with increasing PA levels. The mass of crystals obtained at PA 81.79; 74.23 and 68.59% with a cooling temperature of -12°C is 4.98; 4.09 and 3.18 grams. While at a cooling temperature of -15 °C obtained, the mass of consecutive PA crystals of 5; 4.1 and 3.2 grams. Temperature variations do not have a significant effect on the mass of the crystals obtained. Based on the crystallization curve in Figure 6, it can be seen that this may be because at a temperature of -12 °C the crystal has passed the super solubility curve. Then, if the sample is cooled again at a temperature of -15 °C, the crystal will still be formed but with a small difference.

3.2.2 Effect of Patchouli Alcohol Levels on Yields

From the mass of the crystal obtained, the percentage of crystal yield can be determined at both temperature variations. The results of the crystal yield of patchouli alcohol against distillate PA levels are presented in Figure 7.

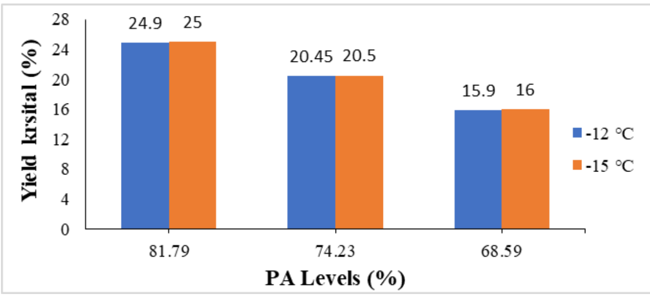
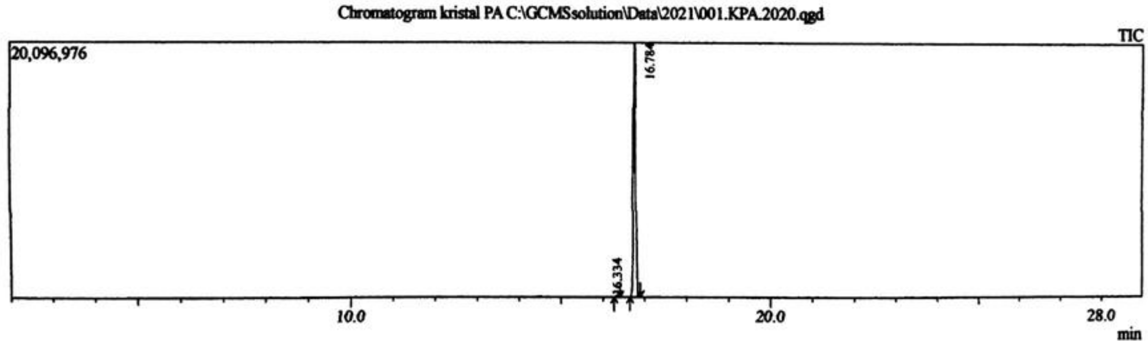


Figure 7. Effects of patchouli alcohol levels on yield obtained



Peak Report TIC

Peak#	R.Time	I.Time	F.Time	Area	Area%	Height	Height%	A/H	Mark	Name
1	16.334	16.270	16.410	317498	0.40	95832	0.48	3.31		.delta.-Guaiene
2	16.784	16.640	16.895	78145755	99.60	20049734	99.52	3.90		Patchouli alcohol
				78463253	100.00	20145566	100.00			

Figure 8. Chromatogram Crystallization of Distillate of Patchouli Oil

From Figure 7, it can be seen that the lower the PA level, the smaller the percentage of crystal yield. Yield obtained from distillate with a PA level of 81.79; 74.23 and 68.59% at -12 °C is 24.9%; 20.45%; and 15.9%. While at a temperature of -15 °C obtained a yield of 25%, 20.5%, and 16%. At temperatures of -15 °C and -12 °C, there is only a slight difference in the mass of distillates obtained so that the difference in yield obtained is also small. A decrease in temperature will induce the rapid formation of crystals to produce higher crystal purity and yield (Mullin 2001).

3.3 Analysis Quality of Crystal

3.3.1 Patchouli Alcohol Levels at Crystal

Patchouli oil quality requirements contained in SNI 06-2385-2006 state that patchouli alcohol levels should not be less than 30%. In the study crystallization of patchouli alcohol with distillates of high-grade patchouli oil with an initial level of PA 81.79; 74.23 and 68.59% resulted in a very high crystal patchouli content of 99.6%. The results of chromatogram Crystal patchouli alcohol can be seen in Figure 8.

3.3.2 Size of Crystal

The study Gotama and Mahfud (2015) mentioned that the speed of stirring at the time of seed addition also affects crystal size distribution. But this study did not use seed addition and only used one variable of stirring speed so that the effect of the comparison of stirring speed or seed addition to the resulting crystal shape could not be seen. SEM testing in this study was only used to find out the average size owned by crystallization results with a variable fixed stirrer rotation speed of 50 rpm.

As illustrated in Figure 9, The crystal form of the SEM result is an average size of 290-600 µm. With it is stated that the size of the resulting patchouli alcohol crystals is quite large and can be used as a perfume raw material or become a seed patchouli alcohol used for further research.

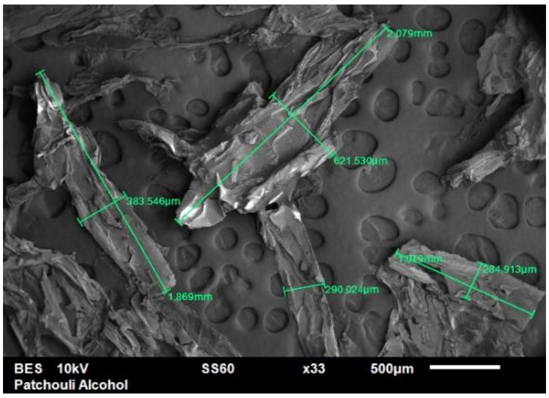


Figure 9. SEM Crystal Patchouli alcohol

4. CONCLUSIONS

Based on the research that has been done, it can be concluded that the lower the operating pressure, the higher the quality of patchouli oil is. The quality of patchouli oil is measured by refractive index parameters, type weights, acid numbers, and patchouli alcohol levels. The lower the cooling temperature in the crystallization process, the more crystal mass is obtained. At the pressure above 1 mbar (-12 °C and -15 °C), the crystal mass obtained respectively is 4.98 and 5 grams. Crystal purity of initial PA level 81.79, 74.23 and 68.59% produce a very high crystalline PA content of 99.6%. The crystal size obtained is an average size of 290-600 µm. With it, it is stated that the size of the resulting patchouli alcohol crystals is quite large and can be used as a raw material for perfumes or become seed patchouli alcohol used for further research.

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