

The application of vacuum redistillation of patchouli oil to improve patchouli alcohol compound

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Abstract. Patchouli oil produced by traditional distillation of patchouli leaves and stems by farmers in Aceh still has low patchouli alcohol compound. In order to increase patchouli alcohol concentration, vacuum redistillation process using packed column was introduced. This research was conducted to fractionate terpene (*alpha-copinene*) from oxygenated hydrocarbon (patchouli alcohol) compound. The operation condition was conducted at two variables that was dependent variable and independent variable. The dependent variable was the 30 cm height distillation packed column, by using raschig ring with 8 mm x 8 mm dimension. And the independent variable was operating temperature 130 °C and 140 °C., vacuum pressure 143,61 mbar, 121,60 mbar and 88,59 mbar and operation time 2 hours, 3 hours and 5 hours. Total of treatments applied in this works were 3 x 3 x 3 or equal to 27 treatments. Patchouli oil used in this research was obtained from Desa Teladan-Lembah Seulawah, Aceh Province. The initial patchouli alcohol compound which analyzed with GC-MS contained 16,02% before treatment applied. After vacuum redistillation process treatment applied patchouli oil concentration increase up to 34,67%. Physico-chemical test of patchouli oil after vacuum redistillation is in accordance with SNI 06-23852006 standard.

1. Introduction

Patchouli plants (*Pogestemoncablin benth*) is one of the important essential oil-producing plants. Among 70 types of volatile oil which are traded in international market, about 9 to 12 essential oils are supplied from Indonesia. Therefore, Indonesia is categorized as an essential oil producing country and a top quality exporter of essential oil. Almost all patchouli plantations in Indonesia are planted by traditional farmers. As one of the largest supplier of patchouli oil in international market, Indonesia contributes 90% of market demand. Most of the patchouli oil products are being utilized in perfume, cosmetic, antiseptic, insecticide and currently aromatherapy industry (Anonymous, 2005). Patchouli oil is an essential oil which has a very important chemical compound namely fixating agent which is used in perfume-based industry, such as perfume, soap, deodorant, etc. This fixative property is caused by its main component of patchouli alcohol ($C_{15}H_{26}O$) belonging to oxygenated terpen. Patchouli oil extracted from patchouli plants (*Pogestemoncablin benth*) by distillation of patchouli leaf [1].

Basically the quality of patchouli oil is strongly influenced by patchouli alcohol content. In general, patchouli oil also contains oxygenated terpen hydrocarbon compounds and other compounds. For certain purposes, in the perfume industry, soaps, cosmetics, and medicines, patchouli oil should be free from terpen compounds due to its ability to the formation of resins in the presence of air. Separation of terpen from patchouli oil would increase its solubility in alcohol and inhibit oxidation



process. The patchouli alcohol component has a relatively high boiling point of 150-160 °C (8mmHg), meanwhile, terpen and sesquiterpene components boiling point is ranging at 150-180 °C and 240-280 °C at atmospheric pressure, respectively [1-3].

Refer to this characteristics, it will be applied terpen separation for patchouli oil purification. The oxygenated compound has a better solubility in alcohol comparable to terpene component and sesquiterpen component. The reduction of terpen component and sesquiterpenes in volatile oil can be conducted by vacuum redistillation, column chromatography and polar solvent extraction. The main problems found in patchouli oil distillation from patchouli leaf which traditionally produced by farmers is still have low patchouli alcohol concentration (Dalimi, 1998). High levels of patchouli alcohol will result in low terpen content resulting in good quality patchouli oil. To produce patchouli oil with high concentration of patchouli alcohol, which is oxygenated terpen compound, it can be done patchouli alcohol component enrichment process with vacuum distillation method with packed column (packed column). In vacuum distillation process of patchouli oil, the fraction with low boiling point (terpen component) produced as a distillate product, whereas the fraction with high boiling point (oxygenated terpen) tends to remain inactive. It is expected that farmers can make changes to patchouli oil distillation equipment from used drums into non-oxidizable metal materials such as stainlesssteel.

The purpose of this research is to separate terpen component, to increase the patchouli alcohol level in patchouli oil by vacuum redistillation method using raschig ring type packed column and to evaluate the effect of temperature, pressure and distillation time to determining parameters quality of produced patchouli oil (refractive index, density, acid number, ester number, and patchouli alcohol level).

2. Materials and methods

2.1. Materials

In this work, patchouli oil obtained from traditional distillation unit in Desa Teladan, Aceh, Potassium hydroxide (KOH, 99,9%: Acros organics), hydrochloric Acid (HCl, 99,9%: Acros organics), phenolphthalein, ethanol (C₂H₅OH, 99,5% Merck), erlenmeyer flasks, beaker glass, measuring cup, burette, picnometer, water bath, spatula, reaction tube, funnel, separator funnel, dropper pipette, volume pipette, scales and refractometer (Reichert Leica). In the vacuum redistillation process the drying and distillation times are determined for 24 hours and 2 hours, respectively. Deionized water was used in whole experiments.

2.2. Methods

A quarter volume of the steam boiler filled with water and heated electrical steam. A 2: 1 proportions of mixed lemongrass leaf and stem put into the distillation kettle equipped with grid where the material was placed. Set the proportion with various density according to the varied. Flow the steam produced by direct contact with raw material to the shelter through the distillate pipe. Drain the oil steam mixture out of the distilled boiler to the condensate through the steam distillation pipe. Accommodated distillate produced on the column of essential oil storage. Repeat the above work by varying the volume of the bed (40%, 60%, 70%, 80%) and sample particle size (3 cm, 5 cm, 8 cm, 12 cm, 15 cm).

3. Results and discussion

3.1. Initial patchouli oli quality before redistillation

The initial patchouli oil analysis results before redistilled can be seen in Table 1.

Table 1. Initial patchouli oil analysis results before redistilled

Parameters	Analysis results (Desa Teladan)
Colour	Brownish yellow
Density	0,9516 (mg/cm ²)
Ester number	17,8605
Acid number	6,0306
Refractive index	1,5074
Solubility in alcohol	1:5
Patchouli alcohol concentration (%)	16,0247

Initial analysis of patchouli oil can be seen that the patchouli alcohol content is very low that is 16.0247% while the standardized in SNI 06-2388-2006 (Table 2.2) is at least 30%. The other parameters already similar to SNI 06-2388-2006 as the color produced brownish yellow. Patchouli oil color can be cleared back with the adsorption process. The higher ester numbers the better (max 20) while the smaller acid number the better (max. 8) quality of patchouli oil. The solubility in alcohol allowed is 1: 10, the more soluble in alcohol the better of its quality. Patchouli oil color after redistillation process changes from yellow brown to dark brown due to part of the light fraction has evaporated so that the concentration of heavy fraction increase. In addition, color changes may also occurred due to a higher heating at low pressure so that the oil near by the kettle wall easily damaged. This also influenced by the burning of oil smells better known as distillate (Ketaren, 1985). If compared with SNI 06-2388-2006, the redistilled patchouli oil color become darker. In SNI 06-2388-2006, patchouli oil is standardized yellow to reddish brown while redistilled patchouli oil is in dark brown.

3.2. Density

The density for each treatment are shown in Figure 1 and Figure 2 for a 2 and 3 hours redistillation time, respectively.

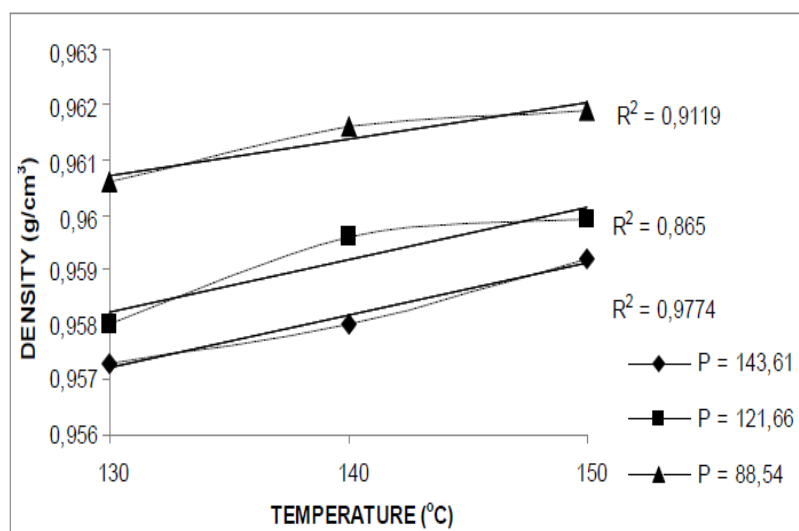


Figure 1. The relationship between temperature (°C) and patchouli oil density which was redistilled at various operating pressures for redistillation time 2 hrs.

Figure 1 shows that as the temperature increase the higher the density of redistilled patchouli oil due to light fraction evaporated. Specific gravity is associated with the heavy fraction of the components contained therein. The larger the weight fraction contained in the oil, the greater the density. Often the heavier type of oxygenated terpen component is greater than that of oxygenated terpenes. The highest density obtained at 88.54 mbar and temperature 1500 °C is 0.9619 g/cm³, and the lowest density is obtained at pressure 143.61 mbar and temperature 1300 °C is of 0.9573 g/cm³. Fig.1 shown that the highest average density value obtained at a pressure of 143.61 mbar is 0.9774 g/cm³.

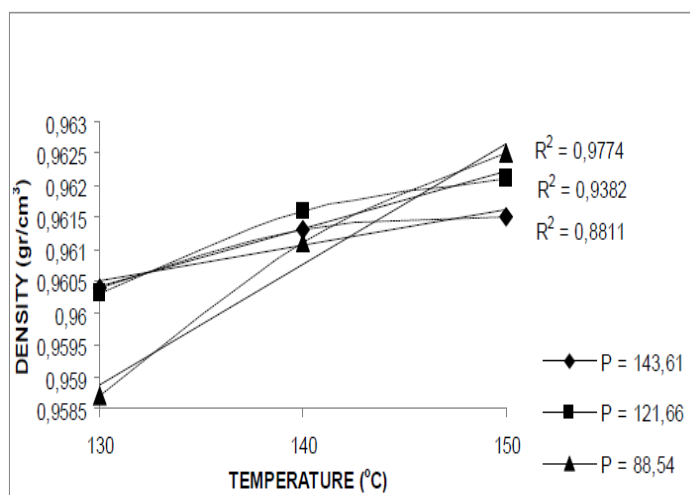


Figure 2. The relationship between temperature (°C) and patchouli oil density which was redistilled at various operating pressures for redistillation time 3 hrs.

Figure 2 shows that the highest density values are obtained under pressure 88,54 mbar and temperature 150 °C that is 0.9625g / cm³, and the lowest value obtained at pressure 88,54 mbar and temperature 130 °C that is 0.9587 g/cm³. The patchouli oil density increased from the initial state from 0.9516g / cm³ to 0.9573 0.9625g / cm³. This increase is due to the process of concentrating heavy fractions on the residue affected by heating and decreasing pressure treatment.

The standardized density in SNI06-2388-2006 is 0.950 0.975 g / cm³. The results showed that the density of patchouli oil from redistillation treatment was within the standard range. If compared between the two process (2 and 3 hours redistillation treatment) it can be seen that the lowest density is 0.9573g / cm³ for 2 hours while the highest density is 0.9625 g / cm³ for 3 hours. On the process of redistilasi for 3 hours obtained the highest average value (mean value) at 88.54. This is due to the longer processing time then the light fraction that evaporates the greater of weight fraction increases and causes the density is also increasing.

The refractive index of essential oils is closely related to the components arranged in the volatile oil produced. Similarly, the specific gravity wherein the essential oil component may affect the refractive index. A long-chain components such as sesquiterpen or oxygen-compounds components involved in distillation process, the density of the essential oil will increase so that the incoming light will be harder to be biased. This causes the oil refractive index increase. The essential oil with a high refractive index is better than low one. The analysis results on refractive index are shown in Figure 3 for redistillation time 2 hours and Figure 4 for redistillation time 3 hours.

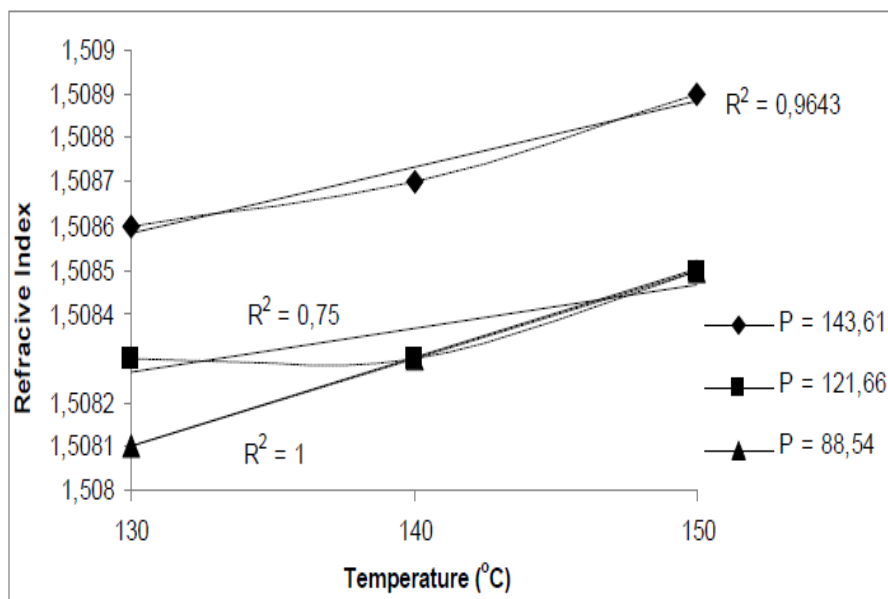


Figure 3. The relationship between temperature (°C) and the index of patchouli oil refraction resulted from redistillation at various operating pressures for a two-hour redistillation time.

If a monochromatic light passes through a medium (A) to a more densely packed medium (B), then the change in velocity and refraction of the light approaches the normal line, or the angle of the incident ray (i_A) greater than the angle of the bias ray (i_B). The comparison of the incident angle of light with this refracted beam angle is called the refractive index. In this study to determine the refractive index of an oil sample used Abbe Refractometer. Figure 3 shows that the highest refractive index values are obtained under pressure 88,54mbar and temperature 1500 °C that is equal to 1,5085 and the lowest refractive index value obtained at pressure 143,61 mbar that is equal to 1,5074.

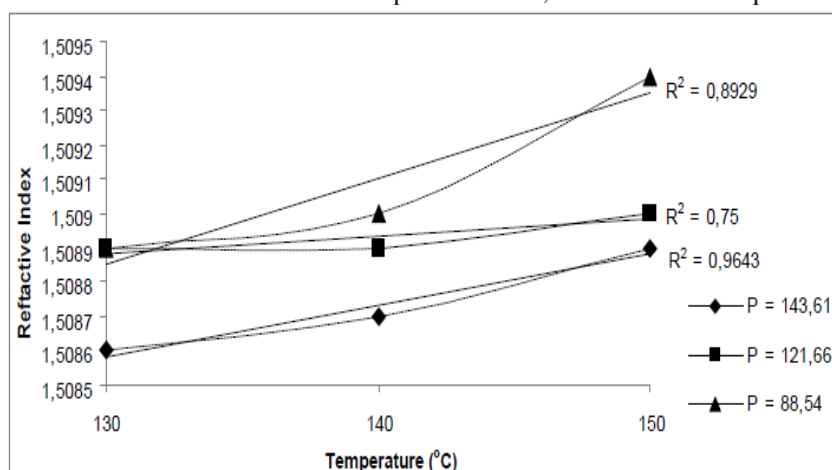


Figure 4. The relationship between temperature (°C) and the index of patchouli oil refraction resulted from redistillation at various operating pressures for a three-hour redistillation time.

Figure 4 shows the highest refractive index values are obtained under pressure 88,54 mbar and temperature 150 °C and the lowest refractive index obtained at pressure 143,61 mbar and temperature 130 °C that is equal to 1,5086, the initial refractive index of 1.5074 increased after the redistillation

process. The refractive index required in SNI 06- 2388-2006 is 1,507 1,515, and the refractive index of patchouli oil of redistillation has been in that range. Comparing Figure 3 with Figure 4, it can be seen that the highest refractive index in the process lasted for 3 hours (Figure 4) and the lowest in the process that lasted for 2 hours (Figure 3). The refractive index on the redistillation process for 2 hours obtained the highest average value at 88.54 and the average value obtained was 1 and the highest average value on the redistillation process for 3 hours was obtained at 143.61 pressure. This is due to the increasing temperature and the lower pressure, the refractive index value will increase. This is because the longer the oil residue process becomes thicker due to the evaporation of mild fraction and water. Index value is also influenced one of them with the water in the oil content. The more the water content, the smaller the value of the bias index. This is because the nature of the water is easy to refract the incoming light.

3.3. Acid Number

Acid numbers show free acid levels in essential oils. The increasing number of acids may affect the quality of essential oils. Namely these acid compounds can change the peculiar smell of essential oils. This may be due to the duration of oil storage and the presence of contact between the volatile oil produced by the light and air around when it is in the oil sample bottle at the time of storage. Most essential oils contain small amounts of naturally occurring free organic acids or those produced from oxidation and hydrolysis ester processes (Ketaren, 1985).

Result of test of acid number for patchouli oil after redistilled for time 2 hours and 3 hours are shown in Figure 5 and Figure 6.

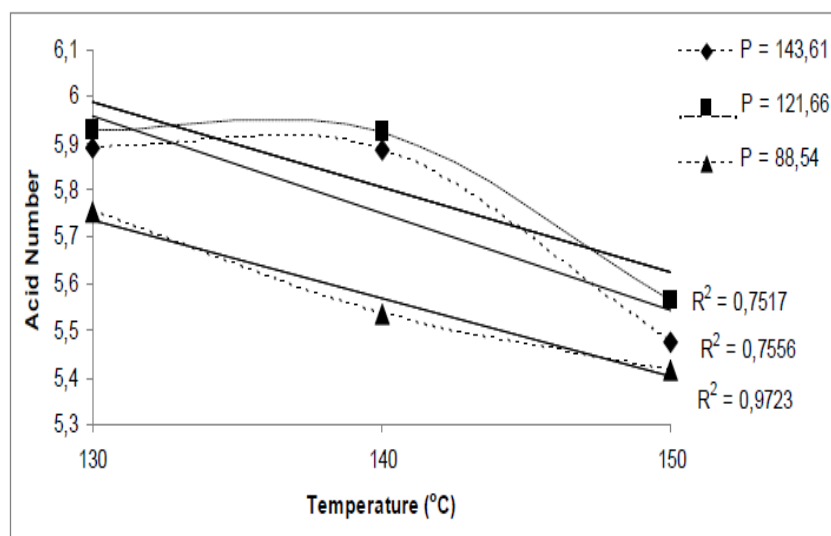


Figure 5. The relationship between temperature (°C) and acid number resulted from redistillation at various operating pressures for a two-hour redistillation time.

Figure 5 shows that the highest acid value is obtained at 121.66 mbar and 1300C at 5.9272 and the lowest acid value obtained at 88.54 mbar is 5,1476 Figure 6 The highest acid number is obtained at 143,61 mbar pressure and temperature 1300 °C that is equal to 5,7818 and the lowest acid number obtained at pressure 121,66 mbar and temperature 1500 °C that is equal to 5,4462. Overall, the patchouli oil acid number before it was redistilled by 6,0306 decreased after the redistillation process. This is due to the distilled patchouli oil that has dried up so that the organic acids vaporize, as suggested by (Guenther 1987) that the oil that has been dried and protected from air and light has a relatively smaller amount of free acid.

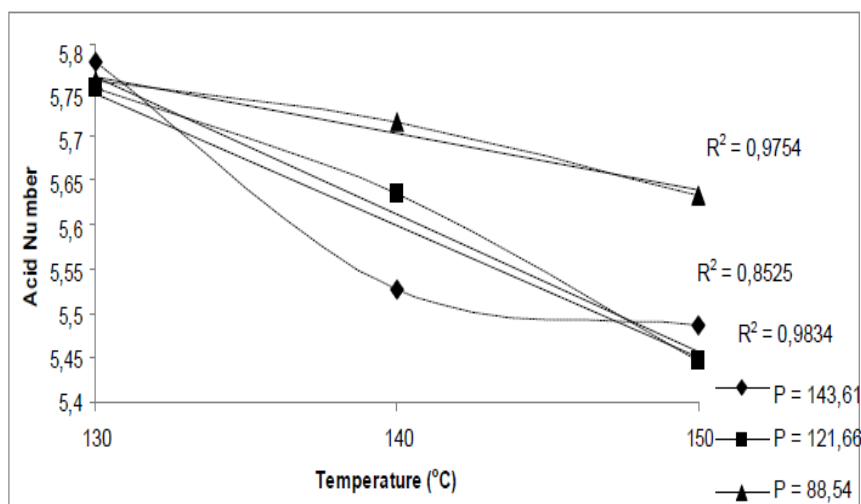


Figure 6. The relationship between temperature (°C) and acid number resulted from redistillation at various operating pressures for a three-hour redistillation time.

When compared between the 2-hour redistillation process (Figure 5) and a 3-hour process (Figure 6) it can be seen that the lowest acid number in the process is 2 hours. The average value obtained in 2-hour redistillation compared to 3 h of redistillation, it can be seen that the average value of 3-hour redistillation is better than the average redistillation value of 2 hours, because with the longer time the oil is likely to oxidize to increase the oil acid number. In SNI 06-2388-2006, it is required that the maximum acid number is 8, and the patchouli oil of the redistillation product is below the prescribed standard.

3.4. *Esther number*

Determination of the amount of esters is very important in determining the value of essential oils. The ester number for the redistilled patchouli oil is shown in Figure 7 for 2 hours refining time and Figure 8 for refining time 3 hours.

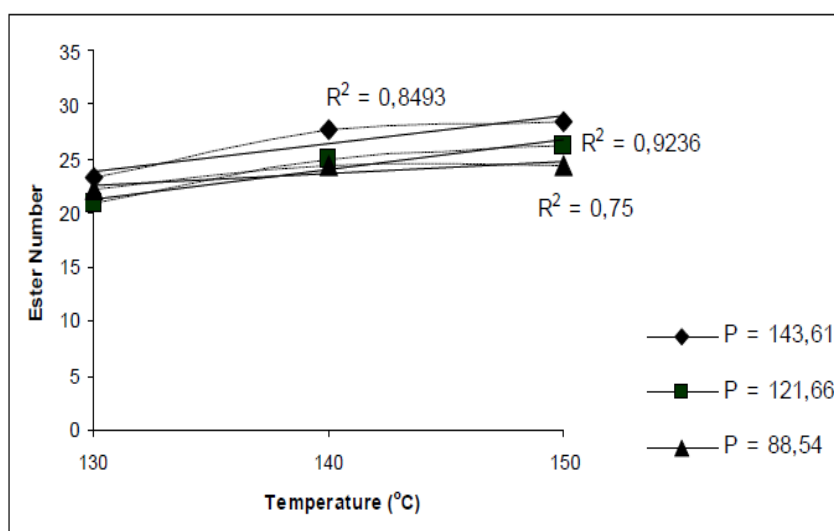


Figure 7. The relationship between temperature (°C) and ester number resulted from redistillation at various operating pressures for a two-hour redistillation time.

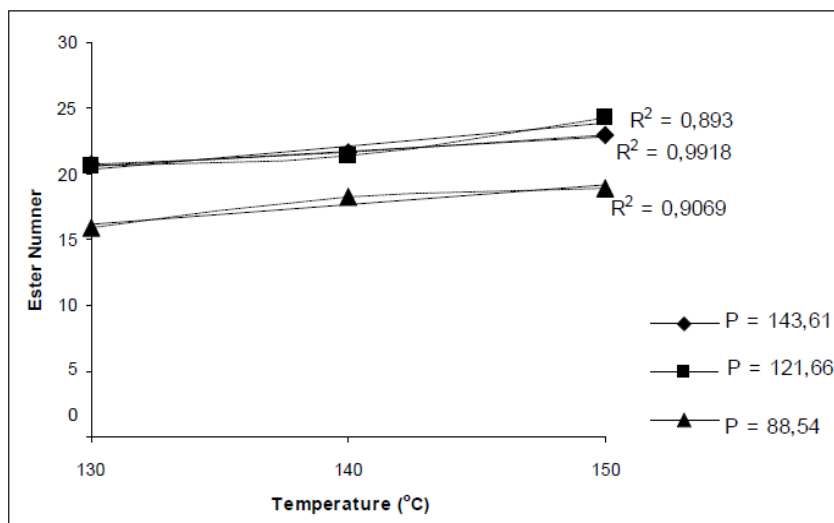


Figure 8. The relationship between temperature (°C) and ester number resulted from redistillation at various operating pressures for a three-hour redistillation time.

Figure 8 The relationship between temperature (°C) and ester number resulted from redistillation at various operating pressures for a three-hour redistillation time Figure 8 shows that at a pressure of 150 mbar, the ester number increases with increasing temperature. The highest ester number is obtained under pressure 121,66 mbar and temperature 1500 °C that is equal to 24,2356 and lowest ester number obtained at pressure 364.44 mbar and temperature 1300 °C yaitu equal to 15,8776. The average value obtained on 2-hour redistillation compared to 3 h of redistillation, it can be seen that the average value of 3-hour redistillation is better than the average redistillation value of 2 hours, because with the longer time it is possible the oxidized oil is greater so it can increase the oil ester number. Overall, the value of patchouli oil ester before it was redistilled was 17.8605 lower than that of patchouli oil after it was redistilled. This is because the ester includes an oxygenated compound with a high boiling point of 2400 2800C at atmospheric pressure as revealed by Guenther (1987) that compounds, phenol, ether and some aromatic aldehyde compounds, sesquiterpen alcohols, esters and so on have a boiling point equal to the boiling point of sesquiterpenes, so that the mild fraction evaporated by the redistillation process increases the fraction of the ester in the residue. SNI 06-2388-2006 requires a maximum estimate of ester 20, but the patchouli oil of redistillation has an ester value higher than 20.

3.5. Solubility in Alcohol

Since many essential oils dissolve in alcohol and are rarely water soluble, the solubility can be readily known by using alcohol at various concentrations. Determining the solubility of the oil depends also on the speed of solubility and the quality of the oil. Oxygenated oils are usually more soluble in alcohol than those rich in terpenes. In lower alcohol concentrations, the terpene fraction will separate as is the case with wax. Based on this, this method is widely used for the manufacture of free oil terpen and sesquiterpen, concentrate and extract. Oil solubility may also change due to the influence of oil age. Unfavorable storage conditions can speed up polymerization. Factors such as light, air and the presence of water usually cause unfavorable effects. The solubility value in patchouli oil alcohol after the redistillation process has increased from 1: 5 to 1: 4. Increased solubility in alcohol occurs because in the process of redistillation, the conventional compound has largely evaporated. P This is because the terpene fraction is somewhat difficult to dissolve in dilute alcohol, while paraffin and sesquiterpen are insoluble at all. Oxygenation compounds generally have better solubility, for example alcohols, aldehydes, ketones and phenols, whereas the ester and phenol ester compounds have smaller solubility values (Guenther, 1987). In SNI 06-2388-2006 it is required that patchouli oil should have a solubility

in alcohol of a maximum of 1: 10 in a clear solution, and patchouli oil of redistilled soluble in alcohol in a ratio of 1: 4.

3.6. Patchouli alcohol content

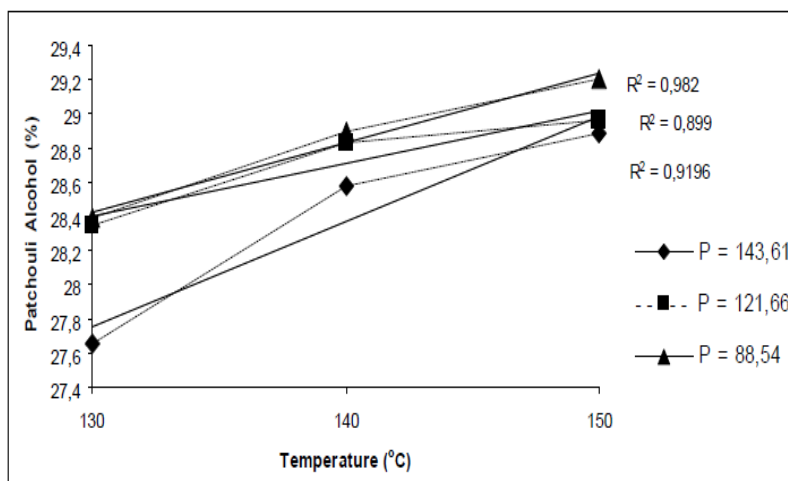


Figure 9. The relationship between temperature (°C) and patchouli alcohol content resulted from redistillation at various operating pressures for a two-hour redistillation time.

Patchouli alcohol is a group of oxygenation compounds is an important component that determines the quality of patchouli oil. The greater the level of patchouli alcohol, the better the quality of patchouli oil produced. Increased levels of patchouli alcohol can be done by the process of redistilasi on the vacuum to vaporize terpen compound. Test results on patchouli alcohol content for patchouli oil resulted from redistillation are shown in Figure 9 for a 2-hour redistillation time and Figure 10 for a 3-hour redistillation time. Figure 9 shows that the level of patchouli alcohol increases with increasing temperature and lower pressure. The highest patchouli alcohol level was obtained at 143,61mbar pressure and temperature 1500C that was equal to 29,12% and the lowest patchouli alcohol level was obtained at 88,54 mbar and temperature 1300C ie by 27.64%.

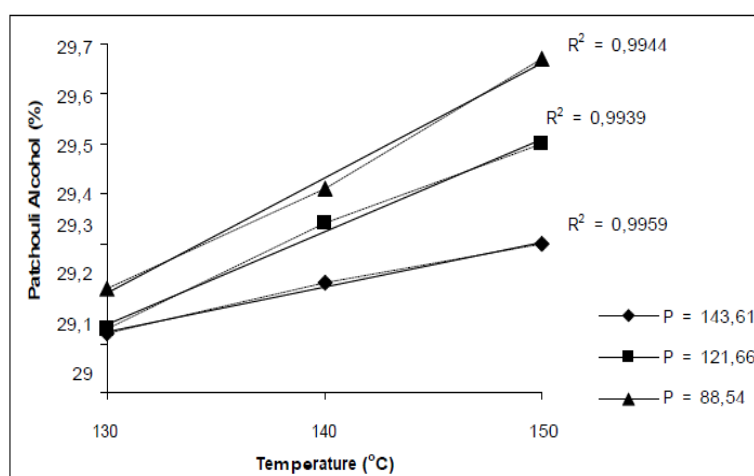


Figure 10. The relationship between temperature (°C) and patchouli alcohol content resulted from redistillation at various operating pressures for a three-hour redistillation time.

Figure 10 shows that patchouli alcohol levels are increasing with increasing temperature and lower pressure. The highest patchouli alcohol content was obtained at 143,61 mbar and temperature 1500 °C that was equal to 29,67% and the lowest patchouli alcohol concentration was obtained at 88,54 mbar and 1300C at 29,13%. Increased levels of patchouli alcohol is quite significant when compared with patchouli alcohol patchouli oil levels before redistilasi. The increase of patchouli alcohol level is above 11,6%, that is from 16,02% to above 27,63%. Comparison of the average value of patchouli alcohol level during redistillation 2 and 3 hours, the average value of redistilasi for 3 hours has a higher value than the average redistilasi 2 hours. This is because the patchouli alcohol level is very influential on pressure, temperature and time. The lower the pressure, the high temperature and the length of time will increase the level of patchouli alcohol in patchouli oil. SNI 06-2388-2006 requires that patchouli oil patchouli oil content is at least 30%, but patchouli oil from redistillation still has not reached the concentration. Patchouli oil from the distillation of the people has a low rpatchouli alcohol so that the process of redistilasi in a vacuum to increase the level of patchouli alcohol. In this research also do the process of redistillation without using the column as the comparison only. The result of patchouli oil redistillation without column Indicates that its physico-chemical properties do not differ greatly from the result of patchouli oil redistillation using the column.

4. Conclusions

From the experimental study the increase concentration of patchouli alcohol in patchouli oil can be conducted by re-distillation process under vacuum conditions. The optimum condition of the re-distillation process is obtained at a pressure of 88.54 mbar and temperature 1500 °C. Patchouli oil result for optimum condition is dark brown, density 0,9611g / cm³, refractive index1,5074, alcohol solubility 1: 4, acid number 5,7129, ester number 18,9073 and patchouli alcohol level is 29,67%. These parameters of optimum conditions is in accordance with SNI 06- 2388-2006 standard.

Acknowledgment

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