

INFLUENCE OF MATERIAL DENSITY AND STEPWISE INCREASE OF PRESSURE AT STEAM DISTILLATION TO THE YIELD AND QUALITY OF CAJUPUT OIL

PENGARUH KEPADATAN BAHAN DAN PENINGKATAN TEKANAN BERTAHAP PADA DESTILASI UAP TERHADAP HASIL DAN MUTU MINYAK KAYU PUTIH

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ABSTRAK

Penyulingan uap pada minyak kayu putih biasa dilakukan dengan tekanan konstan. Penelitian ini membandingkan hasil dan mutu minyak kayu putih dari penyulingan dengan tekanan uap konstan dengan peningkatan tekanan bertahap. Kecepatan bahan selama penyulingan juga harus diperhatikan karena dapat mempengaruhi hasil minyak. Tujuan penelitian ini adalah untuk mengetahui pengaruh kepadatan bahan pada penyulingan uap dengan kenaikan tekanan bertahap terhadap hasil dan mutu minyak. Diharapkan diperoleh hasil yang tinggi dan memenuhi Standar Nasional Indonesia (SNI). Penyulingan dilakukan dengan 3 tingkat kepadatan bahan yaitu 0,20; 0,25, and 0,30 kg/L selama 3,5 jam dengan tekanan awal sebesar 0 bar (pressure gauge) selama 1,5 jam, dilanjutkan dengan 0,5 bar selama 1 jam dan 1 bar pada satu jam terakhir. Rendemen yang diperoleh dari penyulingan tekanan tetap adalah 0,8% dan untuk penyulingan tekanan bertahap berkisar antara 0,79% sampai 0,98%. Kecepatan bahan tidak memberikan pengaruh nyata terhadap rendemen, indeks refraksi dan specific gravity, namun hanya mempengaruhi rotasi optik. Kenaikan tekanan uap bertahap mempengaruhi karakteristik minyak kayu putih. Kadar sineol sebesar 60,48% terdapat pada fraksi dengan tekanan 0 bar, 10,08% pada fraksi dengan tekanan 0,5 bar dan 1,47% pada fraksi dengan tekanan 1 bar. Minyak kayu putih dari penyulingan dengan tekanan konstan hanya mengandung sineol sebesar 35,70%. Komponen utama minyak kayu putih adalah sineol, α -terpineol, trans-kariofilen, viridiflorol, limonen, α -humulen, α -selinen, dan β -selinen.

Kata kunci: penyulingan uap, minyak kayu putih, kepadatan bahan, kenaikan tekanan uap bertahap

ABSTRACT

Steam distillation practices in cajuput oil industry usually use constant pressure. This study compared the yield and quality of cajuput oil from distillation with constant pressure and a stepwise increase of vapour pressure. The material density in distillation also should be considered because it can affect the yield of oil. The objective of this research was to understand the influence of material density on steam distillation with gradual increase of pressure on the yield and quality of cajuput oil produced. It was expected to obtain the highest yield that met specification of Standar Nasional Indonesia (SNI). Distillation carried out with 3 levels of material loading density, i.e. 0.20, 0.25, and 0.30 kg/l for 3.5 hours with the initial pressure gauge of 0 bar for 1.5 hours, then 0.5 bar at 1 hour later, and 1.0 bar at the last hour. The yield produced at distillation was 0.8% for constant pressure and ranged from 0.79% to 0.98% for stepwisely increase pressure. Material density did not have significant effects on yield, refractive index, and specific gravity, but only affected the optical rotation of cajuput oil. Increase in vapour pressure affected the characteristic of cajuput oil. Cineol content was 60.48% on the fraction of 0 bar pressure gauge, 10.08% on the fraction of 0.5 bar pressure gauge, and 1.47% in fraction 1 bar pressure gauge. While cajuput oil from constant pressure contained cineol of only 35.70%. The main components of cajuput oil were cineol, α -terpineol, trans-carryophylene, viridiflorol, limonene, α -humulene, α -selinene, and β -selinene.

Keywords: steam distillation, cajuput oil, material density, stepwise increase vapour pressure

INTRODUCTION

Cajuput oil is an essential oil extracted from fresh leaves and twigs of cajuput tree (kayu putih or white-tea tree, *Melaleuca cajuputi* Powell, Myrtaceae) through water-steam distillation. Cajuput oil, an important forest product in Indonesia, is used for multipurpose home medicine (disinfectant, expectorant, medication for stomach upsets) and as

insecticide (Budiadi *et al.*, 2005). In Java Island, cajuput trees were first planted in 1924, commercial production of the oil began in the 1960s (Subarudi *et al.*, 2005). A high demand of cajuput oil made its industry have important role in the community. National demand of cajuput oil reached about 1,500 tons/year, while national production was only about 577 tons/year (Kusumawardani, 2010).

Cajuput oil is essential oil produced from *M. cajuputi* by distilling the leaves. There were three types of distillation method to produce essential oil, i.e. water distillation; water and steam distillation; and direct steam distillation. Water distillation has many disadvantages including low efficiency and yield and also high risk of hydrolysis reactions (Ketaren, 1985). Water and steam distillation has many advantages. The steam will penetrate smoothly into the tissue materials and the temperature can be maintained up to 100°C. The process is more efficient and effective (shorter time), results in the higher oil yield and better quality of the oil compare to water distillation. Besides that, there were no heat destructions (burnt) on the material (Ketaren, 1985).

Guenther (2006) stated that many essential oils including cajuput oil consist of low and high boiling fractions. Distillation at low pressure (<1 bar) causes steam re-condensed into water on the pile of material. Distillation at high pressure (> 1 bar) can increase the temperature; and the steam penetrate more effectively to the material and accelerate the diffusion process. On the other hand, high temperatures may cause oil hydrolysis and decomposition. Cajuput oil traditionally obtained from water and steam distillation at atmospheric pressure. Cineol, a major compound in cajuput oil, is a compound with low boiling point. Application of a high pressure can increase the risk of losing very volatile fractions and decomposition of unstable compounds. On the other hand, an increase of pressure stepwisely has an advantage to get more oil yield, especially from high boiling point fractions and shorten the distillation time, so that it can reduce fuel cost compare to constant pressure (Lesmayanti, 2004). The aim of this research was to evaluate the effect of increasing pressure stepwisely during distillation process to the yield and quality of cajuput oil fractions and whole oil.

Proper loading of the material quantity also play an important role in the distillation. Loading density of the material need to be considered since it can affect the yield of the oil. Low density of the material allows the hot steam rise up too quick that cause not all of the oil can be vaporized and distilled. On the contrary, too high density may lead the formation of "rat hole" (steam lines) which may allow steam to escape without vaporizing the oil, and also lead "back pressure" phenomena that may decrease the yield. Therefore, this research also evaluated the effect of cajuput leaves and twigs loading density in the distillation chamber to the yield and quality of cajuput oil.

MATERIALS AND METHOD

Materials

M. cajuputi leaves and twigs (diameter < 5 mm) at about 9-10 months old were collected from Wanagama, Gunungkidul, Daerah Istimewa

Yogyakarta-Indonesia. The leaves and twigs were analyzed for moisture content with Aufhauser apparatus. Ethanol 70% and sodium sulphate anhydrous (Na₂SO₄) were also used in this study.

Methods

Determination of Time Span to Increase Pressure Step

Vapor pressure in the boiler was measured with a manometer located on the lid. At the beginning, the distillation was carried out at 0 bar pressure gauge (1 atmospheric pressure). Distillate volume was measured and recorded every 15 minutes to determine the distillation rate. This rate was used to determine the time span for each pressure step in distillation process. When this rate began to decline, the vapor pressure was raised with increment of 0.5 bars. If the rate decreased, then the vapor pressure had to be increased 0.5 bar to maintain the constant rate. The increase of pressure was repeated until there was no oil came out from the condensor. To avoid heat damage, temperature at distillation chamber maintained at 100-150°C, distillates at 40-50°C and condensor water at 50-60°C.

Cajuput Oil Distillation and Analysis

The raw material consists of leaves and twigs of *M. cajuputi* with the ratio of 1:1. The water content was 52.83%. Distillation was carried out using direct steam system. The leaves and twigs were loaded with certain density (0.20, 0.25, 0.30 kg/L) and it was distilled for 3.5 h after first drop of distillate came out. Distillation rate, distillation temperature, and boiler temperature were observed for every 15 minutes. Each fraction of cajuput oil from each pressure step was analyzed its yields and physico-chemical properties. The yield was the weight percentage of cajuput oil to initial weight of leaves and twigs. Then, all the factions from each pressure step (0-1 bar) for each loading density treatment were reconstituted (mixed with certain proportions) and analyzed for physico-chemical properties. A cajuput oil produced with loading density of 0.25 kg/L, and distilled on constant pressure was used as a control.

The measured parameters comprised of specific gravity (picnometer), refractive index (refractometer), optical rotation (polarimeter), and solubility in 70% ethanol. The solubility was measured by put 1 mL of cajuput oil in a glass tube. Drops of ethanol 70% was added and shaken repeatedly until a clear solution was formed. The results were compared to the Indonesian National Standard for cajuput oil SNI 06-3954-2006 (BSN, 2006). Cineol and other volatiles content were analyzed by Gas Chromatography-Mass Spectrometry (GC-MS). Oil samples (0.1 µL) were injected and MS ion chromatograms were recorded

for 30 minutes. Volatile compounds identified based on WILEY7 library and specific gravity.

Yield and quality parameters of cajuput oil fractions at each loading densities at each pressure step were subjected to Analysis of Variance (ANOVA). The experimental design was Single Factor with three levels of leaves loading density (0.20 kg/l, 0.25 kg/l, and 0.30 kg/l) with two replications. Mean differences (α 5%) were determined with Duncan Test.

RESULT AND DISCUSSION

Determination of Time Span at Each Pressure Step

Determination of time span at each step of vapor pressure increase in steam kettle (boiler) was repeated six times (A-F, Table 1). Distillation rate was measured and stopped when no more cajuput oil came out from distillation chamber through a condenser.

Distillation time was varried from 3.5 to 5.0 hours with the data modus of 3.5 hours, so this time span was used for further research. At the beginning, the vapor pressure in the steam kettle was 0 bars, which was the same with atmospheric pressure. The rate of distillate came out from distillation chamber

was 11 L/h/m² or 0.02 mL/minute/cm² of oil and then decreased slowly to less than 5 L/h/m² or 0.008 mL/minute/cm² of oil. Immediately, when it reached this low rates, the pressure of the steam kettle was raised to 0.5 bars. Based on the observation, the time span needed to increase the pressure step from 0 bars to 0.5 bars was 1.5 hours. Distillation rate increased, maintained and after 1.0 hour, it became decreased again gradually. At this point, the pressure level in the steam kettle was re-raised from 0.5 bars to 1 bar and lasted for 1 hour. Increase pressure to 1.5 bars above atmospheric pressure was not required since there was no oil came out, meant that the distillation process was completed.

Effect of Pressure Step and Loading Density

The quality of cajuput oil fractions resulted from distillation for each leaves density with stepwise increase of vapor pressure were evaluated according to SNI 06-3954-2006. The yield of the cajuput oil produced at 0 bar ranged from 0.59-0.73%, at 0.5 bars level ranged from 0.09 – 0.13% and then at 1.0 bar ranged from 0.09 – 0.13% (Figure 1). The value were the mean of two replication.

Table 1. Determination of stepwise increase of pressure

Code	Distillation Time (hours)				Total
	0 bar gauge	0.5 bar gauge	1 bar gauge	1.5 bar gauge	
A	2.0	2.0	1.0	-	5.0
B	1.5	1.0	1.0	-	3.5
C	1.5	1.5	1.0	-	4.0
D	1.5	1.5	1.0	1.0	5.0
E	1.5	1.0	1.0	-	3.5
F	1.5	1.0	1.0	-	3.5

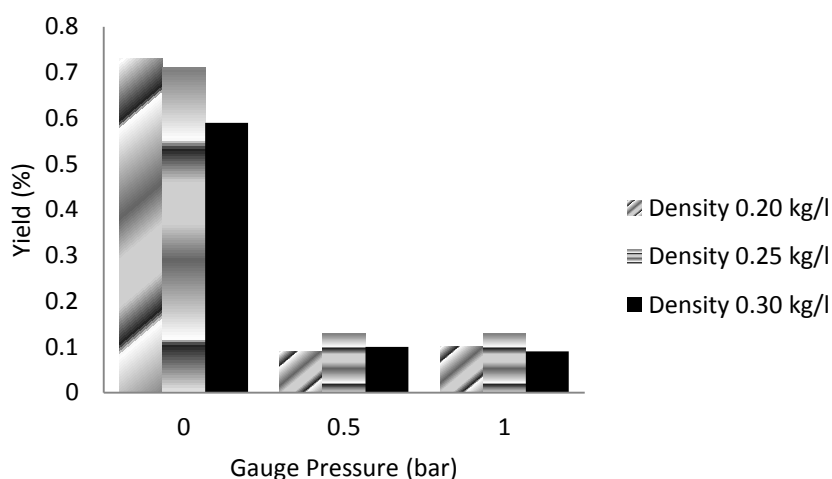


Figure 1. The yield of cajuput oil at different loading density and pressure level

At a pressure of 0 bars, cajuput oil produced was 70%-85% of total oil during distillation (3.5 hours). While at a pressure of 0.5 and 1.0 bar, the oil produced was 7%-15% of total cajuput oil. The oil produces from distillation at 0 bars level contained light fractions with low boiling point. The yields of whole fractions of cajuput oil in this research were 0.78-0.97%. Rusli (2010) said that the yield of cajuput oil was in the range of 0.8-2% depend on the distillation process and plant cultivation. A wide variation in oil yield was found in *M. quinquenervia* trees from Australia and Papua New Guinea. Trees from the South of 25°S have high oil yields (1-3% w/w%), while from the North, were uniformly low (0.1-0.2%) (Ireland *et al.*, 2002).

According to Guenther (2006), the initial fraction of the components with low boiling point will distill first, followed by components with higher one. At the end of distillation, the hydrodiffusion take longer due to the steam need longer time to evaporate higher boiling fractions of the oil. Decrease in yield corresponding with the addition of the distillation time because the gradual increases of pressure were done in sequence.

The color of the cajuput oil determined visually. The oil produced at 0 bar of pressure tend to be clear, while produced at 0.5 bar of pressure have clear greenish-colored. Whereas at a pressure of 1.0 bar, cajuput oil color had a slightly darker green color. The higher pressures of distillation lead the darker color of cajuput oil due to the extracted chlorophyll which had a boiling point at 152°C (Lay, 2003) or possibly a thermal polymerization of terpenoids was occurred.

From the observation, there was odor difference between oil with different pressure level. On the pressure of 0 bars, cajuput oil had a

distinctive odor with fresh and sharp impression. The aroma came from cineol which is the main compound in cajuput oil. Cineol have a camphoraceous, minty, sweet, liquorices, menthol and pine odor. The higher vapor pressure, the fresh and sharp impression decreased due to decreasing amount of the cineol content. At 0.5 and 1.0 bar, the most distilled compounds were trans-carriophyllene which had odor characteristics as terpene-like aroma, musty, green, spicy, woody, fruity and sweet (Hui *et al.*, 2010).

Specific gravity is the ratio of the weight of a given volume of oil to the weight of an equal volume of water measured at a stated temperature and pressure (20°C/20°C). The value of specific gravity of essential oil is quite a narrow range and any deviation from the limit can indicate impurity of the oil. Specific gravity has a positive correlation with the boiling point of components contained in oils. At low pressure, the temperature inside the boiler ranged at 100-110°C. Distilled compounds with low boiling fractions such as monoterpenes and monoterpene-o have a low specific gravity. Increasing in vapor pressure caused the rise of temperature inside the boiler, so that the distilled compounds consisted of higher boiling fractions such as sesquiterpenes and sesquiterpene-o which had high specific gravity. So the higher vapor pressure in distillation, the higher specific gravity of cajuput oil (Figure 2). All fractions met the SNI criteria (0.900-0.930), except 0 bar at 0.2 kg/L density. Low-density of material loading lead to the fast movement of the steam flow rate and uncompleted hydrodiffusion causing fractions with high molecular weight were not penetrated by the steam and were not distilled completely. Therefore, it may decrease specific gravity.

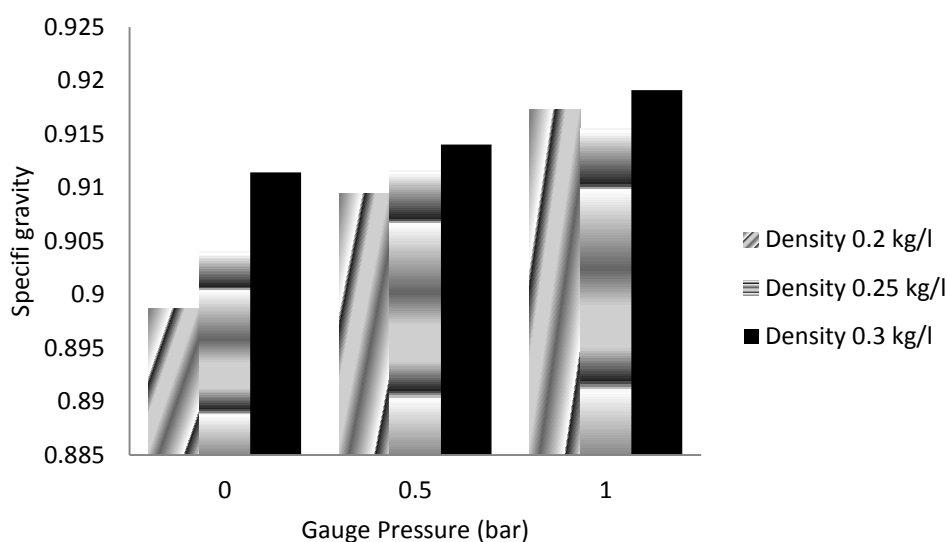


Figure 2. Specific gravity of cajuput oil at different loading density and pressure level

Refractive index is a measure that indicates the refraction of light between oil medium and air. The refraction itself was caused due to the changes in velocities of light beam when passing through two different medium. Refractive Index expresses the ability of whole components of cajuput oil to refract the light that passed and changed the direction at either smaller or bigger angle from normal line. This parameter usually used to early detect adulteration in essential oils. Refractive index was influenced by the length of carbon chain contained in the oil and presence of double bonds. The longer carbon chain and amount of double bonds contained in the oil, then the greater value of its refractive index. Heavy fractions caused the higher density so that the refracted light beam will approached to the normal line (Forma, 1979). Cajuput oil produced from 0 bar was met SNI standards (1.450-1.470) as can be seen at Figure 3. At high pressure, the distilled compounds dominated by heavy fractions that lead to higher density so the refracted laight approached the point of incidence. The higher the vapor pressure used in distillation, then the higher of the refractive index.

The compound is optically active if contained an asymmetric carbon atom, which binds four different atoms or molecules (groups). This optical isomerism caused the difference in electronegativity, which described by the high polarity and chemical binding, resulting in a dipole moment that will rotate plane-polarized to the right (dextrorotatory) and to the left (laevorotatory). The optical rotation of an essential oil was a summation of the optical rotations of its constituents, and was in relation to their proportions within that oil. Cajuput oil contained substances were termed laevorotatory which rotate plane-polarized light to the left. Rizal (2010) stated that the compounds that can rotate plane-polarized light were a compound containing asymmetric carbon atom. Spoelstra (1929) in Guenther (1990) reported the presence of α -terpineol in the cajuput oil that was laevorotatory optically active substance. Only fractions from 0 bar which met the SNI criteria (-4 – 0°). Figure 4 showed that cajuput oil produced from high pressure distillation contained more compounds with dextrorotatory properties. Different optical rotation value indicates different composition of volatile compounds in cajuput oil.

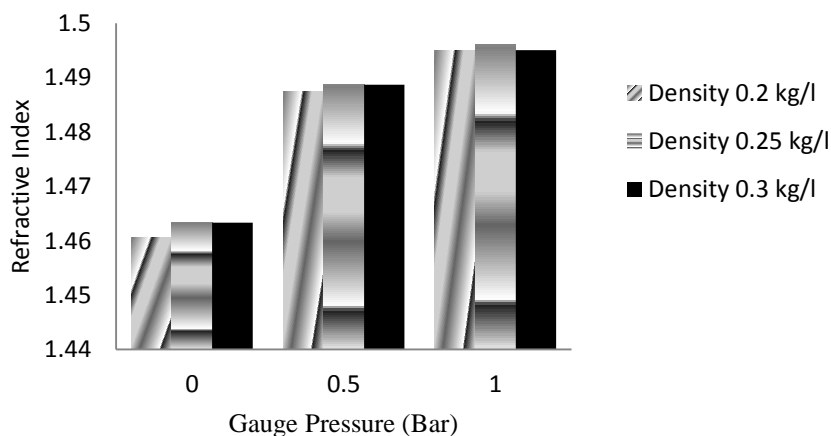


Figure 3. Refractive index of cajuput oil at different loading density and pressure level

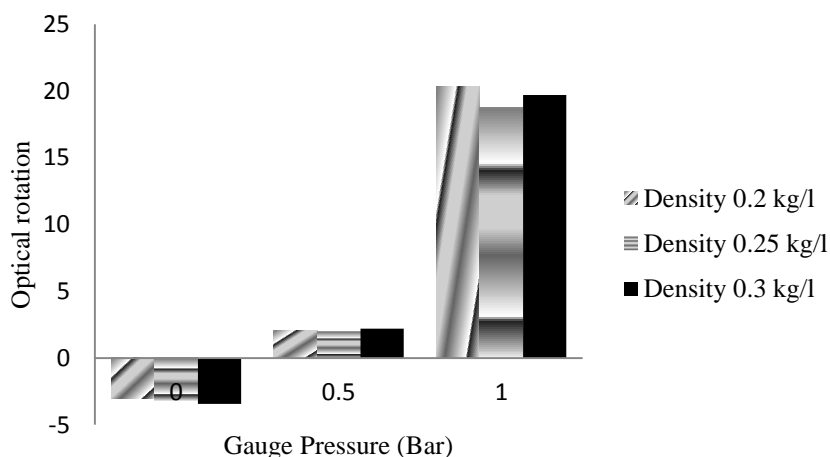


Figure 4. Optical rotation of cajuput oil at different loading density and pressure level

Cajuput oil solubility in ethanol 70% showed different values (Table 2) from every pressure level and increase along with bulk density or loading of cajuput leaves into vessel. All values met the SNI Standards which specified the solubility 1:1 to 1:10. Solubility in ethanol indicates that the fractions contained oxygenated hydrocarbons which are polaric and dissolve completely in 70% ethanol. Oxygenated hydrocarbons included cineol, α -terpineol, veridiflorol, and eudesmol.

Table 2. Cajuput oil solubility in ethanol 70%

Loading density (kg/L)	Pressure (bar)	Solubility in ethanol 70%
0.20	0	1:1
	0.5	1:1
	1.0	1:1
0.25	0	1:1
	0.5	1:5
	1.0	1:10
0.30	0	1:1
	0.5	1:8
	1.0	1:10

Cajuput oil contained a variety of aromatic compounds with specific composition that made the oil has a distinctive aroma. Cineole content in cajuput oil produced in plantations in Java, ranged 67-72% depend on farming systems and site conditions (Budiadi *et al.*, 2005). Volatile compounds were analysed by GC-MS and WILEY7 library. Table 3. shown all volatile compounds detected from each pressure step of distillation (0, 0.5 and 1.0 bar) with loading density of 0.20 kg/L. This treatment was selected because it had high yield (0.92%) and the quality (specific gravity, refractive index, optical rotation and solubility in 70% ethanol) was met with SNI. As a control, it used cajuput oil produced with loading density of 0.25 kg/L at constant pressure distillation.

Fraction from 0 bar of distillation pressure produced cajuput oil containing the highest level of 1,8-cineol (60.48%), 15.93% α -terpineol and 2.36% camphene. For overall, 41 compounds have been identified in this fraction. Fraction from 0.5 bar of distillation pressure produced cajuput oil containing the highest level of trans-carryophyllene as much as 10.298%, 10.08% veridiflorol, 7.03% α -terpineol, 6.56% β -selinene and 6.45% 1,8-cineol. At this fraction, 49 compounds were identified in cajuput oil. Fraction from 1.0 bar of pressure has the highest level of trans-carryophyllene as much as 13.82%, 9.21% veridiflorol, 7.51% α -humulene and 1,8-cineol only 1.47%. This fraction had a total compound of 54 compounds. The control sample of cajuput oil contained 1,8-cineol only as much as 35.70% (less than 50-65% at SNI requirement),

followed by α -terpineol (10.05%), limonene (7.96%), and trans-carryophyllene (5.89%). There were 33 compounds have been identified. From the results, it can be concluded that the main constituents of cajuput oil was cineol and the second was α -terpineol. There were 66 compounds detected in cajuput oil altogether, consisting of 13 monoterpenes, 7 terpene-o, 37 sesquiterpenes and 9 oxygenated sesquiterpenes.

The chemical composition of the leaf oil of cajuput (*M.leucadendra* L.) from Vietnam contains 61 components of which 48 have been identified (Motl *et al.*, 1990). Terpinen-4-ol and terpineol have effective antimicrobial activity (Cuong *et al.*, 1994). Cajuput oil from Cuba has 38 identified components, of which viridiflorol (28.2%) and 1,8-cineole (21.3%) were the major (Pino *et al.*, 2002). While, Ireland *et al.* (2002) stated that cajuput oil produced from *M. quinquenervia* in the region of Australia and Papua New Guinea can be splitted into two chemotype. Chemotype 1 comprised E-nerolidol (74-95%) and linalool (14-30%), and Chemotype 2 contained 1,8-cineole (10-75%), viridiflorol (13-66%), α -terpineol (0.5-14%) and β -caryophyllene (0.5-28%). Wheeler *et al* (2007) also found that two chemical variants or chemotypes exist in *M. quinquenervia* from Florida, each dominated by one sesquiterpene, either E-nerolidol or viridiflorol.

Cajuput oil contained α -pinene, β -pinene, myrcene, α -terpinene, limonene, 1,8-cineol, γ -terpinene, p-cymen, terpinolen, linalool, terpinen-4-ol and α -terpineol. In addition, Doran (1999) said that cajuput oil contained 1,8 cineol, viridiflorol, limonene, trans-carryophyllene, α -humulene, α -terpineol, α -selinene, β -selinene, and carryophyllene oxide. The major compositions of cajuput oils consisted of 49.22% monoterpenes and 46.45% sesquiterpenes, and the rest were hydrocarbons and a diterpene (Kim *et al.*, 2005).

Effect of Loading Density to Quality of Mixed Fractions of Cajuput Oil

Cajuput oil from three levels of leaves loading density (A1-0.20 kg/L, A2-0.25 kg/L, A3-0.30 kg/L) was analyzed for its physico-chemical properties according to SNI. These oils obtained from mixing of all fractions from the same loading density from 0 bar to 1 bar pressure gauge. Cajuput oil produced from distillation with loading density of 0.25 kg/Lat a constant pressure for 3.5 hours was used as a control.

Yield of the cajuput oil ranged from 0.79% to 0.98%, and the loading density does not affect significantly to the yield. However, a trend that the optimum yield obtained from density of 0.25 kg/L was observed. According to Guenther (1972), distillation with higher density of loading material resulted in disruption of the steam flow that can form a vapor line (rat holes).

Table 3. Percentage of volatile compounds detected from GC-MS analysis of cajuput oil from loading density of 0.2 kg/L at each step of vapour pressure

No	Compounds	Quantity (%)			
		0 bars	0.5 bars	1.0 bar	Control
Terpenes					
1	α -Thujene	0.18	-	-	0.37
2	L- α -Pinene	1.91	1.99	0.74	4.27
3	Sabinene	0.16	-	-	-
4	β -Myrcene	1.78	1.21	0.43	1.19
5	β -Pinene	1.93	1.25	0.45	2.98
6	δ -Carene	0.21	-	-	-
7	α -Terpinene	0.50	-	-	0.47
8	β -Cymene	-	1.31	0.49	-
9	1-Limonene	-	4.65	2.16	7.96
10	γ -Terpinene	0.89	0.26	0.24	1.79
11	α -Terpinolene	0.23	0.11	0.13	0.84
12	Isoterpinolene	-	-	0.25	-
13	Camphene	2.36	5.51	3.50	3.32
Terpenes-o					
14	1,8-Cineole	60.48	6.45	1.47	35.70
15	Linalool	0.22	-	-	0.53
16	Cis-Rose oxide	0.09	-	-	-
17	δ -Terpineol	0.45	0.16	-	0.28
18	Terpinene-4-ol	1.35	0.46	0.19	0.97
19	α -Terpineol exo-2-	15.93	7.03	3.56	10.05
20	Hydroxicineole	0.09	-	-	-
Sesquiterpenes					
21	δ -Elemen	-	0.18	-	-
22	Ylangene	0.13	0.99	1.52	0.31
23	α -Copaene	0.10	0.61	0.93	-
24	β -Elemene	0.25	0.50	0.57	0.32
25	α -Gurjunene	-	0.20	0.31	-
26	Trans Caryophyllene	1.97	10.29	13.82	5.89
27	α -Guaiene	-	-	0.20	-
28	Guaia-3,7-diene	-	0.24	0.36	-
29	δ -Gurjunene	-	-	0.24	-
30	γ -Gurjunene	0.14	-	-	-
31	Isolatedene	-	0.32	0.53	-
32	Epizonarene	-	-	0.39	-
33	α -Gurjunene	-	0.33	-	-
34	α -Humulene	1.12	6.10	7.51	3.36
35	Aromadendrene	0.17	0.54	0.71	0.37
Isopropenyl					
36	Dimethyl-Octahydronapthalene	0.18	-	-	-
37	α -Muurolole	-	0.18	0.26	-
38	β -Selinene	-	1.07	1.44	0.57
39	α -Muurolole	0.47	2.95	4.07	1.33
40	β -Maaliene	-	-	4.43	-
41	β -Eudesmene	1.28	6.56	4.30	3.52
42	α -Selinene	1.20	5.01	6.89	2.81
43	Isolatedene	0.22	0.46	1.40	0.63
44	α -Amorphene	-	0.41	0.63	-
45	δ -Cadinene	0.15	0.71	1.09	0.27
46	L-Calamenene	-	0.83	1.12	-
47	γ -Selinene	0.11	1.23	1.77	-
48	Selina-3,7-diene	0.16	0.16	2.14	-
49	Germacrene B	-	0.47	0.63	-
50	Calarene	-	0.27	0.30	-
51	Guaia-5,11-diene	0.21	-	-	0.65

Table 3. Volatile compounds detected from GC-MS analysis of cajuput oil from loading density of 0.2 kg/L at each step of vapour pressure (Continued)

No	Compounds	Quantity (%)			
		0 bars	0.5 bars	1.0 bar	Control
52	Eudesma-3,7-diene	-	1.63	1.53	-
53	β -Guaiene	-	1.82	1.98	-
54	α -Elemene	0.22	-	3.44	0.86
55	δ -Cadinene	-	2.96	-	1.18
56	Juniper camphor	-	-	0.23	-
	Sesquiterpenes-o				
57	Palustrol	-	0.22	0.22	-
	Sesquiterpen-O				
58	Caryophyllene oxide	-	0.36	0.30	-
59	β -Caryophyllene oxide	-	2.37	1.81	-
60	Veridiflorol	1.87	10.08	9.21	3.43
61	Agarospinol	-	0.52	-	-
62	Hinesol	-	-	0.79	-
63	β -Eudesmol	0.31	2.46	3.03	0.99
64	α -Eudesmol	0.41	2.86	3.45	1.31
65	Farnesol	-	0.37	0.45	-
66	Oxo-T-cadinol	-	-	0.32	-

This caused the steam flowed only in the vapor line, resulting lower yield. However, in this study, treatment of the material density didn't give significant effect on the yield of cajuput oil. This is probably due to the density levels which tested were still on the range where the steam can be distributed evenly to all parts of the materials without any significant barriers, so the yield wasn't affected.

Effect of leaves loading density on colour was not significant. All cajuput oils produced have a yellow-greenish in colour that met SNI 06-3954-2006. The colour of cajuput oil was affected by the level of vapour pressure. In the stepwise increase of vapour pressure, the oil produced has a more greenish in colour compared to the oil produced with constant pressure. High level of vapour pressure will produce oil which consists of more diversified and numerous compounds. Each essential oil has its own specific aroma. According to Ketaren (1985), the odors of the essential oils came from oxygenated hydrocarbon compounds. Cajuput oil contained some oxygenated hydrocarbons such as cineol, α -terpineol, veridiflorol, and eudesmol.

Specific gravity is affected by the chemical components contained in the oil that are used to evaluate the oil purity. So if the value is too high or too low, it indicated the presence of other compounds that shouldn't present in cajuput oil. The more heavy fractions contained in the oil, then the higher its specific gravity. All cajuput oil produced has a specific gravity that met SNI criteria. From the analysis, increasing in material density didn't influence significantly on the specific gravity of the oil. Material loading density also did not affect significantly to the refractive index. Loading density affected optical rotation of the oil significantly. Increase of loading density caused greater optical

rotation due to much more of optically active substances were distilled (Figure 5).

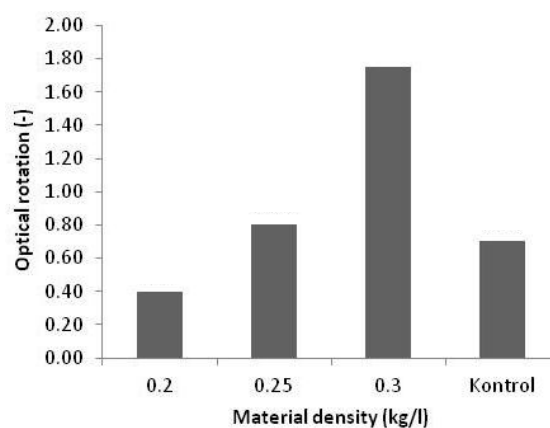


Figure 5. Optical rotation of cajuput oil at different material loading density

Essential oils have ability to dissolve in ethanol at a certain ratio. According to Guenther (2006), the chemical components contained in the essential oil will determine the oil solubility in ethanol. Oxygenated hydrocarbon compounds more soluble in ethanol compared with terpene hydrocarbons such as oxygenated hydrocarbons. Oxygenated hydrocarbon compounds are relatively more polar than terpenes compounds. The best solubility in 70% ethanol was cajuput oil produced from distillation with material density of 0.20 kg/L which ratio of oil and ethanol was 1:1. This oil may contain higher oxygenated hydrocarbon compounds than the other samples which had ratio of 1:5, including control.

CONCLUSIONS AND RECOMMENDATION

Conclusions

The oil yields produced from stepwisely increase pressure distillation were slightly increase ranged from 0.79% to 0.98%, while control at constant pressure was 0.8%. Increase in vapour pressure affected the composition of cajuput oil compounds. Cineol content was 60.48% on the fraction of 0 bar pressure gauge, 10.08% on the fraction of 0.5 bar pressure gauge, and 1.47% in fraction 1 bar pressure gauge. While cajuput oil from constant pressure contains cineol only 35.70%. The main constituents of cajuput oil were cineol, α -terpineol, trans-caryophyllene, viridiflorol, limonene, α -humulene, α -selinene, and β -selinene. Loading density had no significant effects on yield, refractive index, and specific gravity, but affected the optical rotation of cajuput oil.

Recommendation

Stepwise increase of pressure can fractionate cajuput essential oils effectively, especially to increase cineol content up to 60%. The application of cineol rich fractions for cajuput candy or aromatherapy products were interesting to study.

REFERENCES

- Budiadi, Hiroaki I, Sigit S, Yoichi K. 2005. Variation in kayu putih (*Melaleuca leucadendron* LINN) oil quality under different farming systems in Java, Indonesia. *Euras J For Res.* 8 (1): 15-20.
- [BSN] Badan Standarisasi Nasional Indonesia. 2006. Minyak kayu putih SNI 06-3954-2006. Jakarta
- Cuong ND, Xuyen TT, Motl O, Stansky K, Presslova J, Jedlickova Z. 1994. Antibacterial properties of Vietnamese Cajuput oil. *J Essent Oil Res.* 6: 63-67.
- Doran JC. 1999. *Melaleuca cajuputi* Powell. Plant Resources of South-East Asia. No. 19: Essential-oils plants. Prosea Foundation, Bogor, Indonesia. pp. 126-131.
- Forma MW. 1979. *Physical Properties of Fats and Fatty Acids In: D. Swern (ed). Baileys Industrial Oil and Fat Products.* New York: John Wiley and Son.
- Guenther E. 1972. *Minyak Atsiri Jilid IV.* Ketaren S, penerjemah. Jakarta: Universitas Indonesia Press. Terjemahan dari *Essential Oil.*
- Guenther E. 1990. *Minyak Atsiri Jilid IV B.* Ketaren S, penerjemah. Jakarta: Universitas Indonesia Press. Terjemahan dari *Essential Oil.*
- Guenther E. 2006. *Minyak Atsiri Jilid I.* Ketaren S, penerjemah. Jakarta: Universitas Indonesia Press. Terjemahan dari *Essential Oil.*
- Hui YH, Feng C, and Leo MLN. 2010. *Handbook of Fruit and Vegetable Flavors.* New York: John Wiley and Sons.
- Ireland BF, Hibbert DB, Goldsack RJ, Doran JC, Brophy JJ. 2007. Chemical variation in the leaf essential oil of *Melaleuca quinquenervia* (Cav.) S.T. Blake. *Biochem Sys Ecol.*30: 457-470.
- Ketaren S. 1985. *Pengantar Teknologi Minyak Atsiri.* Jakarta: Balai Pustaka.
- Kim JH, Liu KH, Yoon Y, Sornnuwat Y, Kitirattrakarn TT, Anantachoke C. 2005. Essential Leaf Oils from *Melaleuca cajuputi*. *Acta Hort.* (ISHS) 680:65-72
- Kusumawardani PK. 2010. Analisis tingkat kepuasan pelanggan terhadap produk minyak kayu putih produksi perum perhutani [Skripsi]. Bogor: Fakultas Teknologi Pertanian, IPB.
- Lay MB. 2003. *Chlorella, The Ultimate Green Food.* Detroit Lakes, BL Publications
- Motl O, Hodačová J and Ubik K. 1990. Composition of Vietnamese Cajuput essential oil. *Flavour Fragr. J.* 5: 39-42.
- Pino J, Bello A, Urquiola A, Aquero J, Marbot R. 2002. Chemical composition of Cajuput oil (*Melaleuca leucadendra* L.) from Cuba. *J Essential Oil Res.* 14(1):10-11.
- Rizal S. 2010. Kajian proses penyulingan minyak nilam menggunakan sistem distilasi air [skripsi]. Bogor: Fakultas Teknologi Pertanian, Institut Pertanian Bogor.
- Rusli MS. 2010. *Sukses Memproduksi Minyak Atsiri.* PT Jakarta: AgroMedia Pustaka.
- Subarudi TD, Suwardi E, dan Setiyono H. 2005. Social and economic aspects of Cajuput oil industry in Indonesia: A rapid assessment in Java and NTT [Research Report]. Center for Social and Economic Research on Forestry. Forestry Research and Development Agency (FORDA), Ministry of Forestry. Indonesia
- Wheeler GS, Pratt PD, Giblin-Davis RM, Ordnung KM. 2007. Intraspecific variation of *Melaleuca quinquenervia* leaf oils in naturalized range in Florida, the Carribean and Hawaii. *Biochem Sys and Ecol.* 35: 489-500.