Effect of Distillation Tank Density and Storage Time on the Quality and Chemical Composition of Cajuput Oil

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Abstract

Cajuput oil is a commodity of non-timber forest product which is needed and potential to be developed in Indonesia. Therefore, further research on the factors of production and post-production are needed to produce optimum quality. In this study, the leaves of cajuput (*Melaleuca cajuputi* Powell) were distillated by water-steam distillation. This study evaluated effects of distillation tank density (60%, 70%, and 80%) and oil storage time (0 month, 1 month, 2 months, and 3 months) on physicochemical properties and chemical compositions of cajuput oils. The results showed that cajuput oils had a specific gravity of 0.915~0.923; optical rotation of (-2,10°) ~ (-1,20°); refractive index of 1.463~1.464; solubility in alcohol 1 : 1; cineole contents of 61.18~76.24%; clear to yellowish clear color; distinctive smell of cajuput and in accordance with SNI 06-3954-2006. The GC-MS analysis identified 24 of chemical components on the cajuput oils with main components were 1,8-cineole, α-pinene, and β-caryophylene. Tank density 70% to 80% with the oil storage time up to 3 months still gives the optimum quality and chemical compositions.

Keywords : cajuput oil, distillation tank density, storage time, physico-chemical properties, chemical composition.

Introduction

Cajuput oil is one of potential essential oil in Indonesia. Several plants species which producing cajuput oil and planted in Indonesia are *Melaleuca cajuputi* Powell and M. *leucadendron* Linn. (Widiyanto and Siarudin 2013). Other species which is being develop are M. *minor* Smith. and M. *viridiflora* Gartn. (Guenther 1987).

Cajuput oil production from natural forest in Maluku Archipelago with area of approximately 120,000 ha reached 196 ton/year and after economy crisis in Indonesia, it decreased to 100 ton/year. In Java, the states (Perum Perhutani) manage cajuput plants in the area of 18,000 ha with oil productivity of 300 ton/year, meanwhile Forestry Service of Yogyakarta Province manage cajuput plants in the area of 4,000 ha with oil productivity of 50 ton/year (Kartikawati and Rimbawanto 2014).

In fact, state production is insufficient to meet domestic demand. Cajuput oil packing industries or pharmacy industries information according to Kartikawati and Rimbawanto (2014) reported that state cajuput oil need reach up to 1,500 ton/year while the yearly supply is only 400 ton. The shortage of more than 1,000 ton is closed by importing eucalypt oil from China. Those problems could be overcome if the plants parts and processing factory is revitalized (Kartikawati and Rimbawanto 2014). There are some factors in oil processing which ascertain the quantity (yield) and quality of cajuput oil products. Factors influencing yield i.e. climate and soil, harvesting month, plants age, plants species, plants spacing, leaves condition, and oil processing. Factors influencing oil qualities i.e. plants species, leaves storaging method, tank filling which is related to leaves density in the tank, oil taking steps, and oil storage after processing (Kasmudjo 2011).

Related to the tank filling method especially cajuput leaves density in the distillation tank is if the leaves density in the tank is too high, it would prevent the steamed water to evaporate in the distillation process and makes the leaves wet which escalating the hydrolysis process and decreasing the oil quality. Moreover, the blocked steamed water would push to make way out and resulting in making uneven flow in the entire leaves. Some oils would stay in the leaves. Thus, the oil quality would decrease (Sumadiwangsa and Silitonga 1977). Leaves filling about half and a quarter of tank volume and adding the branch up to 20% would maintain the production of oil with high quality (cineole content up to 50%). In the factor of cajuput oil storage after processing, it could evaporate in room temperature without decomposing, being in the group of volatile oil (Kasmudjo 2011). Cajuput oil storage would evaporate the main components inside the oil (Guenther 1987). Turek and Stintzing (2012) analyze that essential oil composition is easily changed in the isolated oil storage. Volatile oil is susceptible to conversion and degradation reaction of oxidation process and polymerization caused by temperature, lights, and oxygen availability. Main components of cajuput oil are 1,8-cineolee, sesquiterpene alcohol globulol, viridiflorol, and spathulenol, and also minor components such as: limonene, β-caryophyllene, humulene, viridiflorene, α-terpineol, α dan β-selinene, and caryophyllene oxide (Brophy and Doran 1996). Quality decreasing caused by chemical changing would influence the pharmacological function of the cajuput oil (Turek and Stintzing 2012). Volatile oil which contains high monoterpene or oxide (cineole) has the storage time of 1~3 years (Anonymous 2014).

Due to those concerns, this study evaluated variations of distillation tank density 60%, 70%, and 80% of tank volume, in the oil storage time after processing of 0 month, 1 month, 2 months, and 3 months to obtained optimal yield of cajuput oil quality and compositions.

Materials and Methods

Preparation and Essential Oil Samples

Fresh leaves of cajuput (*M. cajuputi* Powell) were obtained from RPH Menggoran Playen, Gunung Kidul, Yogyakarta, Indonesia. Leaves and terminal twigs were taken about 1 kg, and then the leaves are separated to the branches. Branches used should have the length of about 25 cm and diameter of 0.4 mm. Average proportion of leaves and branches obtained from this method was 78% : 22%. In order to get the optimum proportion of leaves and branches (80% : 20%), leaves mass was added by 2% and branches mass was reduced by 2%. Distillation tank capacity used in this research was 10 kg. Thus, to make 60% of tank density, it was filled up to 6 kg, to make 70% of tank density, it was filled up to 7 kg, and to make 60% of tank density, it was filled up to 8 kg. Water-steam distillation method was used in this study. Cajuput oils were produced put in dark glass bottle and storage for 3 months at close storage with room temperature (±28°C) and humidity of 84%.

Physicochemical Properties

Physico-chemcial properties testing of cajuput oils were done based on SNI 06-3954-2006. Analyses determined color, odor, specific gravity at 20°C, refractive index at 20°C, solubility index in 70% alcohol, and optical rotation. Oil color was analyzed based on visual observation, and odor was evaluated by the direct smell of paper strip containing the oil.

Optical rotation of oil was measured by disk polarimeter (WGX-4, Shanghai Benson Instrument Co. Ltd, Shanghai, China). Optical rotation is expressed in degrees of circumference (º). Optical rotation *dextro* is a positive sign (+) and optical rotation *levo* is a negative sign (-).

Specific gravity was measured by pycnometer based on the weight ratio of oil and water in the same volume at the same temperature. This test used 5 ml volume of pycnometer. The empty pycnometer weighed (m), then it was filled with distilled water (avoiding any air bubbles) and weighed (m_1). Then pycnometer was washed with ethanol and subsequently diethyl ether, and dried. This pycnometer filled with oil and weighed (m_2). The specific gravity of oil was obtained using the following equation:

$$d^{20} = d^t + 0.0007 \times (t-20)$$

where d^{20} is specific gravity at 20°C, t is ambient temperature (ºC), 0.0007 is correction factor.

$$d^t = (m_2 - m)/(m_1 - m)$$

where d^t is specific gravity at ambient temperature (t°C), m (g) is weight of empty pycnometer, m_1 (g) is weight of pycnometer contained water at t°C, m_2 (g) is weight of pycnometer contained oil at t°C.

Refractive index was determined by handy refractometer (N-3000e, Atago Co., Ltd, Tokyo, Japan). The refractive index was calculated by following equation:

$$n_D = n^t_D + 0.0004\ (t-20)$$

where n_D is index value at 20°C, n^t_D is index value at ambient temperature (t°C), 0.0004 is correction factor.

Solubility in 70% alcohol was estimated based on the volume ratio of oil to 70% alcohol. One ml of oil is put in 10 ml volumetric glass and 1 ml of 70% alcohol was added, then solution becomes clear after mixing well. If solution is not clear, 1 ml of 70% alcohol is further added until clear solution is obtained. The results are expressed as follows:

Solubility in 70% alcohol = (1 ml of oil) : (ml of 70% alcohol added)

GC-MS Analysis

Cajuput oil chemical composition were analyzed by GCMS-QP2010S SHIMADZU with the specification as follows: AGILENT HP 5MS column (0.25 mm id x 30 m, film thickness of 0.25 nm), carrying gas is Helium, and ionization EI 70 eV, oven column temperature 70°C, injection temperature 310°C, injection mode is splitless, sampling time 0.20 minutes, electric current control mode was pressure of 13.7 kPa, total current is 60 mL / minute, current column: 0.50 mL / minute, with linear velocity of 25.9 cm / second. Oven temperature was programmed from 70°C (5 minutes on hold) to 300°C (19 minutes on hold), and from 100~230°C every 15°C/minute raised was hold on 5 minutes. Chemical compositions were identified base on WILEY 229 data base library and were compared with some literatures.

Statistical Analysis

All experiments were replicated three times, and the data were averaged. The results were tested by two-way ANOVA. Significant differences between means were determined by Honestly Significant Different (HSD) test. Value of $P < 0.05$ were considered statistically significant.

Results and Discussion

Physicochemical Properties

Physicochemical properties of cajuput oil were evaluated including color, smell, specific gravity, optical rotation, refractive index, alcohol solubility ratio, cineole content, and were presented in the Table 1. Physicochemical variance analysis of Cajuput oil in this study were presented in Table 2.

Table 1. Physicochemical properties of Cajuput Oils

Sample	Specific Gravity	Optical rotation(°)	Refractive Index	Alcohol solubility ratio	Cineole Content (%)	Color	Smell
K_1L_0	0.917	-2.10	1.463	1 : 1	67.68	Clear – Yellowish Clear	Original Cajuput Smell
K_1L_1	0.921	-1.70	1.464	1 : 1	67.08		
K_1L_2	0.921	-1.37	1.464	1 : 1	76.24		
K_1L_3	0.922	-1.23	1.464	1 : 1	62.34		
K_2L_0	0.915	-2.23	1.463	1 : 1	65.95		
K_2L_1	0.920	-1.83	1.463	1 : 1	66.37		
K_2L_2	0.920	-1.33	1.463	1 : 1	74.75		
K_2L_3	0.922	-1.40	1.464	1 : 1	61.18		
K_3L_0	0.920	-2.00	1.463	1 : 1	68.80		
K_3L_1	0.922	-1.37	1.464	1 : 1	68.86		
K_3L_2	0.923	-1.20	1.463	1 : 1	76.03		
K_3L_3	0.923	-0.90	1.464	1 : 1	64.24		
Total Mean	0.921	-1.56	1.464	1 : 1	68.29		

Notes:
K_1: Tank density 60%, K_2: Tank density 70%, K_3: Tank density 80%, L_0: Oil Storage Time 0 month, L_1: Oil Storage Time 1 month, L_2 : Oil Storage Time 2 months, L_3: Oil Storage Time 3 month.

Table 2. Variance analysis of Cajuput Oil

Variance	Specific density	Optical rotation (°)	Refractive index	Solubility in 70% alcohol
Tank density (K)	6.41 **	1.60 ^{ns}	0.68 ^{ns}	- ^{ns}
Oil Storage Time (L)	10.74 **	7.16 **	0.67 ^{ns}	- ^{ns}
Interaction (K*L)	0.71 ^{ns}	0.12 ^{ns}	0.09 ^{ns}	- ^{ns}

Notes:
ns: not significant *:significant ** : very significant

Color and Smell

Color test result according to tank density and storage time entirely showed the same clear-yellowish color and is defined as good. This condition is relevant to the standard of SNI that is clear to yellowish green. Essential oils in this study have smell same with original cajuput smell. This condition is relevant to the standard of SNI that is original cajuput smell (Anonymous 2006).

Specific Gravity

Specific gravity of the cajuput oil is varied by the different factors of tank density and oil storage time about 0.915~0.924 with the mean of 0.921 and it is defined as high and good. The average value of this specific gravity is relevant to the quality standard of Indonesia National Standard (SNI) which is about 0.900~0.930 (Anonymous 2006).

According to the factor of leaves density in the distillation tank of 60%, 70%, and 80%, the average value of specific gravity are 0.920; 0.919; and 0.922 respectively. The density of 80% results the highest specific gravity and followed by 60% and 70%. It is presumed that there is a difference of the weighed fraction of chemical component contents in the cajuput oil at the different factors of tank density. The more materials are processed, the more weighed fraction of chemical composition contents is contained in the material. Specific gravity is defined by the amount of oil components. The more long chain components such as sesquiterpen (C_{15}) and oxygen based components or oxygenated hydrocarbon is processed, the higher medium density or the specific gravity of the volatile oil processed (Ariyani *et al*. 2008). Oil of the distillation tank density of 80% contains the highest sesquiterpen and oxygenated hydrocarbon contents i.e. 92.83%, while on the density of 60% is 91.86%, and on the density of 70% is 90,90%. There are 16 chemical components in cajuput oil which are included in the group of sesquiterpen and oxygenated hydrocarbon, i.e.: butanoic acid, benzene, 1,8-cineole, 4-terpineol, α-terpineol, α-terpinyl acetat, viridiflorol, caryophyllene oxide, nerolidol, β-caryophyllene, α-humulene, β-gurjunene, β-selinene, α-selinene, α-caryophyllene, and β-elemene.

According to the factor of oil storage time of 0, 1, 2, and 3 months, the average value of specific gravity are 0.917; 0.921; 0.922; and 0.922, respectively. Specific gravity value is also increase gradually from the storage time of 0~3 months. It is presumed that the presence of oxidation and polymerization cause the average value of cajuput oil specific gravity raise from the storage time of 0~3 months. Oxidation of volatile oil could decrease the amount of chemical components in the oil (Ketaren 1985). Essential oil composition is easily changing in the process and the isolated oil storage, where the external factors such as temperature, lights, and oxygen availability influence the changing of chemical composition process (Turek and Stintzing 2012). The amount of hydrocarbon component which has low saturation point is easily decreased in the room temperature storage (Najafian 2014). It is presumed that long components and components which has low saturation point is more resistant of those chemical reaction. The more long chained components such as *sesquiterpen* (C_{15}) and/or oxygen based components, the higher medium density or specific grafity of volatile oil would be (Ariyani *et al.* 2008). In this cajuput oil process, sesquiterpene and *oxygenated hydrocarbon* contents tends to increase on the storage time of 0 to 3 months by 90.03%, 91.53%, 93.77%, and 92.13% respectively. Terpene containing oil if is stored at the long period of time would form a certain resin or usually called as resinification or polymerization (Ketaren 1985). The resinous content presumes to take a certain part in the increasing of specific gravity. The higher specific gravity value due to the increasing of the resinous content would decrease the quality of the oil.

Variance analysis showed that the factors of tank density difference and oil storage time are significant, while the interaction is non-significant. The different variables of tank density and storage time factors influence the specific gravity, but if those factors are combined would not significantly influence the specific gravity. Storage time would be more important than tank density because ANOVAs value of the storage time is higher. Thus, it is important to pay attention to the oil storage time of cajuput oil processing to produce the optimal specific gravity.

Optical Rotation

Optical rotation of the cajuput oil is varied by the different factors of tank density and oil storage time about (-2.50°) ~ (-0.50°) with the average of -1.56° and is defines as high and good. The average value of optical rotation is relevant to the Indonesia National Standard i.e. (-4°) ~ (0°) (Anonymous 2006).

According to the factor of leaves density in the distillation tank of 60%, 70%, and 80%, the average value of optical rotation are -1.60°; -1.70°; dan -1.37°. The density of 80% results the highest specific gravity and followed by 60% and 70%. It is presumed that the value of optical rotation is linear to the value of specific gravity. The higher specific gravity, the higher optical rotation would be. The value of optical rotation is relevant to the value of the specific gravity according to the factor of tank density.

According to the factor of oil storage time of 0, 1, 2, and 3 months, the average value of optical rotation are -2.11°; -1.63°; -1.30°; and -1.18°, respectively or generally tends to increase. The value of optical rotation tends to increase by the increasing of specific gravity according to the factor of oil storage time of 0~3 months. The increasing of specific gravity is also increasing the value of optical rotation.

Variance analysis showed that the factor of oil storage time is very significant to optical rotation, while the factor of tank density and the interaction of both factors are non-significant. It is showed that the factor of oil storage time gives higher significant influence to optical rotation as one of cajuput oil quality value.

Refractive Index

Refractive index of the cajuput oil is varied by the different factors of tank density and oil storage time about 1.462~1.465 with the average of 1.464 and is defined as high and good. The average value of refractive index is relevant to the Indonesia National Standard i.e. 1.450~1.470 (Anonymous 2006).

According to the factor of leaves density in the distillation tank of 60%, 70%, and 80%, the average value of refractive index are nearly the same, i.e. 1.464; 1.463; dan 1.463 respectively. It is presumed that the value of refractive index is linear to the value of specific gravity and optical rotation. Oil which have high refractive index is usually also have high density (Effendi and Widjanarko 2014). The more long chain components such as sesquiterpen (C_{15}) and oxygen based components is processed, the higher medium density or the specific gravity of the volatile oil processed, and the incoming light would be more difficult to refract. This would cause the higher refraction index of the oil (Ariyani *et al.* 2008).

According to the factor of oil storage time of 0, 1, 2, and 3 months, the average value of refractive index are 1.463; 1.464; 1.464; dan 1.464, respectively and tends to increase slightly. It is presumed that resinification reaction cause the average value of specific gravity increase from the storage time of 0~3 months. The more long chain components such as sesquiterpen (C_{15}) and oxygen based components is processed, the higher medium density or the specific gravity of the volatile oil processed, and the incoming light would be more difficult to refract. This would cause the higher refraction index of the oil (Ariyani *et al.* 2008).

Variance analysis showed that the factor of tank density, oil storage time and both factors interaction is non-significant on the value of refractive index in the oil. It is showed that the factor of tank density and oil storage time give non-significant influence to the value of refractive index.

Solubility in 70% Alcohol

Alcohol solubility ratio of the cajuput oil by the different factors of tank density and oil storage time is entirely same for about 1 : 1 with clear condition which is defined as very good. The alcohol solubility ratio is relevant with the quality standard of SNI i.e. (1 : 1) ~ (1 : 10) (Anonymous 2006).

According to the factor of leaves density in the distillation tank of 60%, 70%, and 80%, the value of alcohol solubility content are entirely the same 1 : 1. Feasibility of the oil to soluble in the alcohol is presumably caused by the high presence of oxygenated hydrocarbon components in the cajuput oil even though being processed at the different density, which is about 82.37~ 83.39%. It is relevant to the statement of Ketaren (1985), where oxygenated hydrocarbon components have higher solubility in liquid alcohol.

According to the factor of oil storage time of 0, 1, 2, and 3 months, the average value of alcohol solubility ratio are entirely the same for about 1 : 1. It is presumed that oil storage time factor is non-significantly influence the value of alcohol solubility ratio and the oil is evenly good. Oxygenated hydrocarbon components in cajuput oil is still high and not much change in the storage time of 0~3 months, and showed that the oil solubility in water is still good.

Cineole Content

Cineole content of the cajuput oil is varied by the different factors of tank density and oil storage time about 61~76% with the average of 68%. The average value of optical rotation is even higher than SNI i.e. 50~65% (Anonymous 2006).

According to the factor of leaves density in the distillation tank of 60%, 70%, and 80%, the average value of cineole content are 68.34%, 67.06%, and 69.48% respectively. According to Kasmudjo (2011) by filling half and a quarter (75%) volume of the distillation tank and adding the branches up to 20%, would produce the still high cineole content in the oil. It is presumed that by the tank density of 60%, 70%, and 80%, would show the still high cineole content at the main parameter of cajuput oil quality and relevant to the standard even if it is varied. Tank density of 80% produces the highest average of cineole content, while the lowest is 70%. It is presumed that it is caused by the higher oxidation occur at the tank density of 70% due to the leaves condition during distillation process or in the wetter storage condition. Oxidation of volatile oil could decrease the amount of chemical content in the oil (Ketaren 1985).

Table 3. T-test cineole content of Cajuput Oil related to tank density.

Variable	Mean	t test
K_1	0.68 ± 0.06	5.76 *
K_2	0.67 ± 0.06	
K_1	0.68 ± 0.06	-2.37 ns
K_3	0.70 ± 0.05	
K_2	0.67 ± 0.06	-6.08**
K_3	0.70 ± 0.05	

Notes:
K_1: Tank density 60%, K_2: Tank density 70%, K_3: Tank density 80

Table 4. T-test cineole content Cajuput Oil according to oil storage time.

Variable	Mean	t test
L_0	0.68 ± 0.01	0.134 ns
L_1	0.67 ± 0,01	
L_0	0.68 ± 0,01	-16.78 **
L_2	0.76 ± 0,01	
L_0	0.68 ± 0.01	20.98 **
L_3	0.63 ± 0.02	
L_1	0.67 ± 0.01	-14.28 **
L_2	0.76 ± 0.01	
L_1	0.67 ± 0.01	27.95 **
L_3	0.63 ± 0.02	
L_2	0.76 ± 0.01	19.97 **
L_3	0.63 ± 0.02	

Notes:
L_0: Oil Storage Time 0 month, L_1: Oil Storage Time 1 month, L_2: Oil Storage Time 2 months, L_3: Oil Storage Time 3 months.

T-test result at the factor of oil storage time to cineole content in Table 4 showed that storage time of 0 to 1 month is not significant, while storage time of 0 to 2, 0 to 3, 1 to 2, 1 to 3, and 2 to 3 is very significant.

According to the factor of oil storage time of 0, 1, 2, and 3 months, the average value of cineole content are 68%, 67%, 76% and 63% respectively. The highest cineole content is found at the storage time of 2 months which is 76%, while the lowest is at 3 months which is 63%. It is presumed that oxidation process in cajuput oil cause the cineole content in the oil become unstable in the storage time of 0 to 3 months. According to Ketaren (1985) oxidation process of volatile oil both cause smell changing and decreasing the amount of chemical components in the oil. Oxidation process in volatile oil mainly occur in the double chain reaction such as in 1,8-cineole ($C_{10}H_{18}O$), where 1,8-cineole is included in the group of *oxygenated hydrocarbon* component which contained terpene unit. In this research, the oil is kept in the minimum light room, but at the condition of room temperature and the storage cup still have room for air, oxygen (O_2) in the air would increase the occurrence of oxidation reaction and decreasing the oil quality. This occurrence is relevant to Najafian (2014) statement that the decreasing quality occurs higher in the volatile oil of *Melissa officinalis* during the storage at the room temperature to the temperature of 4°C and -20°C. Composition changing and the physical-chemical properties of volatile oil is generally more significant in the half-filled cup than only a little or none air room (Turek and Stintzing 2013).

T-test on the tank density factor to cineole content at the Table 3 showed that between the density of 70% and 80% is very significant, between the density of 60% and 70% in significant, while between the density of 60% and 80% is non-significant.

Table 5. Chemical composition of Cajuput Oil (%).

Compound	Chemical Formula	Retention Time	K_1				K_2				K_3				Mean
			L_0	L_1	L_2	L_3	L_0	L_1	L_2	L_3	L_0	L_1	L_2	L_3	
Butanoic acid	$C_6H_{12}O_2$	3.458	nd	0.41	nd	0.50	nd	0.66	0.17	0.91	nd	0.35	nd	0.39	0.28
α-thujene	$C_{10}H_{16}$	6.395	nd	nd	nd	Nd	nd	nd	0.18	nd	nd	nd	nd	nd	0.02
β-ocimene	$C_{10}H_{16}$	6.554	nd	nd	nd	3.01	3.93	nd	nd	3.07	2.75	2.87	nd	2.60	1.52
α-pinene	$C_{10}H_{16}$	7.002	4.02	3.15	1.87	Nd	nd	3.48	2.45	nd	nd	nd	2.11	nd	1.42
β-pinene	$C_{10}H_{16}$	8.337	3.05	2.56	1.66	2.66	3.11	2.80	2.05	2.73	2.38	2.80	1.87	2.27	2.50
β-myrcene	$C_{10}H_{16}$	8.705	2.91	2.09	1.24	2.04	2.68	2.33	1.54	2.16	2.05	2.21	1.47	2.09	2.07
p-cimene	$C_{10}H_{16}$	9.456	nd	nd	0.37	Nd	nd	nd	0.43	nd	nd	nd	nd	nd	0.07
Benzene	$C_{10}H_{18}O$	9.658	nd	nd	nd	Nd	nd	0.64	nd	nd	nd	nd	nd	nd	0.05
1.8-cineole	$C_{10}H_{18}O$	9.962	67.68	67.08	76.24	63.34	65.95	66.37	74.75	61.18	68.00	68.86	76.03	64.24	68.23
γ-terpinen	$C_{10}H_{16}$	10.587	0.86	0.45	0.3	0.34	1.33	0.69	0.57	0.61	0.86	nd	0.32	nd	0.53
α-terpinolene	$C_{10}H_{16}$	11.600	nd	nd	nd	Nd	nd	nd	0.25	nd	nd	nd	nd	nd	0.02
4-terpineol	$C_{10}H_{18}O$	14.740	0.86	0.77	0.57	0.84	0.85	0.85	0.62	0.90	nd	0.88	0.63	0.87	0.72
α-terpineol	$C_{10}H_{18}O$	15.037	10.94	10.47	7.95	11.97	9.91	9.17	6.91	10.9	10.48	10.69	8.35	12.17	9.99
α-terpinylacetat	$C_{12}H_{20}O_2$	19.859	1.87	2.69	1.89	3.42	3.23	4.49	3.3	6.23	1.80	2.91	2.17	3.76	3.15
β-caryophyllene	$C_{15}H_{24}$	21.989	6.15	6.12	3.30	9.65	7.13	6.36	3.26	7.03	8.45	6.65	3.39	8.12	6.30
α-humulene	$C_{15}H_{24}$	23.142	nd	nd	nd	Nd	nd	nd	nd	nd	nd	1.78	nd	nd	0.15
β-selinene	$C_{15}H_{24}$	22.918	nd	1.88	1.41	2.45	nd	1.77	1.35	2.24	nd	nd	1.44	2.37	1.24
α-caryophyllene	$C_{15}H_{24}$	23.322	1.68	nd	nd	Nd	1.89	nd	nd	nd	2.05	nd	nd	nd	0.47
β-elemene	$C_{15}H_{24}$	23.802	nd	1.73	1.38	Nd	nd	nd	0.86	1.36	nd	nd	0.99	0.50	0.57
β-gurjunene	$C_{15}H_{24}$	23.882	nd	nd	nd	Nd	nd	nd	nd	nd	1.18	nd	nd	nd	0.10
α-selinene	$C_{15}H_{24}$	24.027	nd	nd	1.25	Nd	nd	nd	0.91	nd	nd	nd	0.85	nd	0.25
Nerolidol	$C_{15}H_{26}O$	26.447	nd	nd	nd	0.78	nd	nd	nd	0.67	nd	nd	nd	nd	0.17
Viridiflorol	$C_{15}H_{26}O$	26.527	nd	0.61	0.56	Nd	nd	nd	0.41	nd	nd	nd	0.37	nd	0.16
Caryophyllene oxide	$C_{15}H_{24}O$	26.765	nd	nd	nd	Nd	nd	0.40	nd	nd	nd	nd	nd	nd	0.03

Notes:

nd: not detected, K_1: Tank density 60% , K_2: Tank density 70%, K_3: Tank density 80%, L_1 : Oil Storage Time 1 month, L_2 : Oil Storage Time 2 months, L_3 : Oil Storage Time 3 months

Table 6. Oxygenated hydrocarbon content in Cajuput Oil according to the factors of tank density and storage time (%).

Tank density	Storage time				Mean
	L_0	L_1	L_2	L_3	
K_1	81.35	82.03	87.21	79.84	82.61
K_2	79.94	82.58	86.16	80.79	82.37
K_3	80.28	83.69	87.55	82.03	83.39
Mean	80.52	82.77	86.97	80.89	82.79

Notes:
K_1: Tank density 60%, K_2: Tank density 70%, K_3: Tank density 80%, L_0: Oil Storage Time 0 month,
L_1: Oil Storage Time 1 month, L_2 : Oil Storage Time 2 months, L_3: Oil Storage Time 3 months.

Table 7. Hydrocarbon content in Cajuput Oil according to the factors of tank density and oil storage time (%).

Tank density	Storage time				Mean
	L_0	L_1	L_2	L_3	
K_1	18.66	17.98	12.78	20.15	17.39
K_2	20.05	17.43	13.85	19.20	17.63
K_3	19.72	16.31	12.44	17.95	16.61
Mean	19.48	17.24	13.02	19.10	17.21

Notes:
K_1: Tank density 60%, K_2: Tank density 70%, K_3: Tank density 80%, L_0: Oil Storage Time 0 month,
L_1: Oil Storage Time 1 month, L_2 : Oil Storage Time 2 months, L_3: Oil Storage Time 3 months.

Chemical Composition

GC-MS analysis identified 24 components in cajuput oil (Table 5). The most abundant component is 1,8-cineole with the average of 68.23%, while the lesser is α-thujene with the average of 0.02%. There are three main chemical components of cajuput oil i.e.: 1,8-cineole (68.23%), α-terpineol (9.99%), and β-caryophyllene (6.30%).

Chemical components of cajuput oil are divided into two groups, oxygenated hydrocarbon and hydrocarbon. There are 9 chemical components which are included into oxygenated hydrocarbon (C, H, dan O) group, i.e.: butanoic acid, benzene, 1,8-cineole, 4-terpineol, α-terpineol, α-terpinyl acetat, viridiflorol, caryophyllene oxide, and nerolidol (Table 6).

There are 15 chemical components which are included into hydrocarbon $(C_5H_8)_n$ groups, i.e.: α-thujene, β-ocimene, α-pinene, β-pinene, β-myrcene, p-cimene, γ-terpinen, α-terpinolene, β-caryophyllene, α-humulene, β-gurjunene, β-selinene, α-selinene, α-caryophyllene, dan β-elemene (Table 7). The relationship of hydrocarbon group and oxygenated hydrocarbon group is contradictory. Oxygenated hydrocarbon components influence the quality of cajuput oil include specific gravity, optical rotation, refractive index, and alcohol solubility ratio.

According to the factor of leaves density in the distillation tank of 60%, 70%, and 80%, the average value of oxygenated hydrocarbon are 82.61%; 82.37%; and 83.39% respectively. It is presumed that at the density of 80%, material condition is still relatively porous even though the material process is the most abundant. Thus, the distillation process would run optimally and the weighed fraction chemical components such as oxygenated hydrocarbon and sesquiterpene are abundant in the oil, while the lowest is at the density of 70%. It is presumed that that is caused by the higher oxidation process occur in the density of 70% due to the leaves condition in the process and the storage is wetter. Oxidation process in volatile oil could decrease the amount of chemical components of volatile oil (Ketaren 1985).

According to the factor of oil storage time of 0, 1, 2, and 3 months, the average value of hydrocarbon content are 19.48%; 17.24%; 13.02%; and 19.10% respectively. Hydrocarbon contents tend to decrease in the storage time of 0 to 3 months. It is presumed to be caused by the occurrence of oxidation process during the storage. Oxidation process in volatile oil could decrease the amount of chemical components of volatile oil (Ketaren 1985). This is relevant to the research by Rowshan *et al.* (2013) to the volatile oil of *Thymus daenensis* which showed that at the room temperature, hydrocarbon components proportion with low saturation point such as α-pinene, α-terpinene, myrcene, γ-terpinene, dan p-cymene decrease after storage time up to 3 months. The amount of hydrocarbon components with low saturation point would easily and highly decrease at the room temperature. This phenomenon is cause by evaporation, oxidation, and other unwanted changing in volatile oil during the storage (Najafian 2014). After 3 months storage time, oxygenated hydrocarbon components are started to decrease, and this presumed to be caused by oxidation. These components are more resistant to oxidation than hydrocarbon components which already decerase by the storage time of 0 month. This is relevant to the statement of Ketaren (1985) that oxygenated hydrocarbon components are more resistant and stable to oxidation and resinification, while the hydrocarbon components are the contrary.

Conclusions

Cajuput oils in this study have quality in accordance with SNI 06-3954-2006. These oils consist of 24 compounds with three major compounds of 1,8 - cineol , α - terpineol , and β - caryophyllene . Oxygenated hydrocarbon compound contents was between 82.37~83.39%, and hydrocarbons between 16.6~17.63%. Distillation tank density of 70% to 80% gives optimum yield and quality, whether storage time of cajuput oils until 3 months still have good chemical composition.

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