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Effect of crushing of clove bud (*Syzygium aromaticum)* and distillation rate on main constituents of the essential oil

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Abstract
The essential oil from clove bud (*Syzygium aromaticum*) was extracted by hydrodistillation. The main compounds of the essential oil were eugenol, caryophyllene and eugenyl acetate. Distillation process was prepared with whole clove bud and crushed clove bud. Composition of eugenol, β-caryophyllene and eugenyl acetate for whole clove bud were 77.53%, 5.90 %, 12.07 % respectively and crushed clove bud were 72.40%, 12.61%, and 9.59 % respectively. The particle size influenced composition of eugenol, caryophyllene and eugenyl acetate for variation of time in distillation process. Component of eugenol in whole clove bud increased from 78.16 % to 79.93%, caryophyllene decreased from 7.22% to 5.17% and eugenyl acetat decreased from 8.69% to 7.65%. Effect of crushing of clove bud made difference of composition. Component of eugenol in crushed clove bud increased from 44.54 % to 74.78%, caryophyllene decreased from 44.92% to 12.83% and eugenyl acetate increased from 1.07% to 6.8%.

Keywords: *Syzygium aromaticum*, essential oil, chemical composition.

1. Introduction
The essential oil as the essence of the plant's fragrance is also called volatile oil because they evaporate quickly when exposed to the air at room temperatures. Essential oils are used to give flavour to foods and drinks and as fragrance in the food and cosmetics industries, where numerous herbal and spice ingredients are components in the manufacture of skin creams, lip balms, shampoos, soaps and perfumes [1].

An essential oil is defined internationally as the product obtained by hydrodistillation, steam distillation or dry distillation of a plant or some part of it. They are aromatic oily liquids, volatile, characterized by strong odour, rarely coloured, and generally with a lower density than of water [2]. Volatile oils are complex mixture of compounds consisting of monoterpenes, sesquiterpenes, and their oxygenated derivatives (alcohols, aldehydes, esters, ketones, phenols, and menthol [3].

Clove, native to the small islands of Maluku in Eastern Indonesia also known as the "Spice Islands", has been traded from one end of the world to the others, being highly sought after commodity in medieval Europe for medicinal and culinary purposes [4].

Clove (*Syzygium aromaticum*) is an aromatic herb that has many useful purposes. The aroma of clove is pleasant yet spice and has some medicinal purposes as well and it tastes good in certain dishes like spice cake. Clove likes to grow in hot tropical climates area as of Indonesia. Part of the clove that is commonly used is the flower buds of clove. The aromatic oils of the clove can increase blood circulation and raise person's temperature slightly. The oil of the cloves have been known to stimulate and disintect body as it travels through the body. In foods and beverages, clove is used in toothpaste, soaps, cosmetics, perfumes, and cigarettes [5, 6, 7].

Clove is used to help digestion, prevent vomiting in pregnancy and has inhibitory effect on histamine production [8]. Clove oil will stop the pain of a toothache when dropped into a cavity [9].

The clove tree is an evergreen tropical plant, which flowers twice a year. Cloves are the unopened buds and harvested when the outer green leaves have change from green to yellow pink [10]. Clove oil is obtained from distillation of the flowers, stems, and leaves of the clove tree (*eugenia aromatica*). Clove essential oils have been analyzed by GC-MS [11].

The most important constituents of clove is the phenylpropene eugenol which gives this spice

its pengent, distinctive aroma. Eugenol makes up 70% to 90% of the essential oil and 15 % of the dry weight of clove buds [12, 13]. Several constituents of clove have been identified, mainly are eugenol, eugenyl acetate, beta-caryophyllene, 2-heptanone, alpha- humulene, methyl salicylate, isoeugenol, methyl eugenol, phenyl propanoides, dehydrodieugenol, kaemferol, gallic acid, and oleanolic acid [14].

The objective of this research was to study effect of crushing Indonesian clove bud on chemical composition and oil distillation rate. However there are no previous references in literature about such kind of comparison results on identification and evaluation of crushing effect for Indonesian clove bud oil.

2. Material and methods

2.1. Plant collection

Dry clove bud type of java was gained from collection of clove bud from PT Indesso Aroma, Cileungsi, West Java, Indonesia.

2.2. Sampel preparation

The levels of moisture content from dry clove bud was checked by moisture analyzer and the result was 9.86%. Grinding process is carried out using a blender machine until its particle size.

2.3. Extraction ef essential oil

2.3.1 Identification of clove oil composition

50 gram of crushed clove bud was extracted using clevenger distillation apparatus in ratio of sample and water was 1:5. Hydrodistillation process was running for 24 hours.

2.3.2 Distillation rate and chemical composition.

50 gram of whole clove bud and crushed clove bud were carried out in hydrodistillation using clevenger apparatus. The oil was identified by GC-MS for distillation from one hour to seven hours.

2.4. Analysis of the oil

Clove bud were analyzed by GC-MS Agilent HP7890 GC System, Agilent 5975C series GC MSD. GC 7890 with FID detector, FID heater 275 ^{o}C, column agilent HP-1 methyl siloxane (30 m x 250 µm x 0.25 µm film thickness), carrier gas Helium with pressure 2.0244 psi; inlet: split ratio 500:1, total flow 53.1 mL/min, septum purge flow 3 mL/min, gas saver on 20 mL/min after 2 minutes. Oven temperature was programmed from 100 ^{o}C for 10 minutes, then 5 ^{o}C/min to 200 ^{o}C for 0 minutes, then 10 oC/min for 10 min, total running time was 45 min. MS Acquisition parameters: EMV mode was relative, relative voltage was 0, resulting EM voltage was 2506; scan parameters: low mass 30.0, high mass 500.0, threshold 50; MS zones: MS Source 250 ^{o}C and MS quadrupole 200 ^{o}C, ionization voltage 70 eV, MSD transfer line 300 ^{o}C. Compound identification was done by comparing the Wiley 2008-NIST library and P. Adam data of the peaks with those reported in literature, mass spectra of the peaks with literature data.

3. Result and Discussion

The oil yield of whole clove bud and crushed clove bud were 14.50% and 14.45% w/w, respectively. Differences of particle size did not give significant differences in the yield of clove oil on 24 hour of distillation. The chemical compositions of the oil were identified by GC-MS and the result as shown in Table 1.

Table 1: Chemical constituents of whole clove bud and crushed clove bud

No	RI	Chemical composition	Percentage	
			whole clove bud	crushed clove bud
1	853	2 –heptanone	0,01 ± 0,00	-
2	920	5-methylfuran-2-carbaldehyde	0,01 ±0,00	-
3	938	6-methylhept-5-en-2-one	0,01 ±0,00	-
4	984	Ethyl hexanoate	0,01±0,00	-
5	1036	Benzyl alcohol	0,01±0,00	-
6	1052	Methyl heptyl ketone	0,01 ±0,01	0,01±0,00
7	1082	Linalool	0,01 ±0,00	0,01±0,00
8	1160	Benzyl acetate	0,01 ±0,00	0,01±0,01
9	1203	4-allylphenol	0,20 ±0,15	0,21±0,06
10	1221	α – copaene	0,11 ±0,05	0,41±0,08
11	1228	3,7-dimethylocta-2,6-dien-1-ol	0,01 ±0,00	0,02±0,02
12	1281	Methyl salicylate	0,06 ±0,01	0,05±0,00
13	1344	α – cubebene	0,05 ±0,08	0,11±0,02
14	1361	Eugenol methyl ether	0,02 ±0,00	0,01±0,00
15	1386	Alloaromadendrene	0,02 ±0,00	0,04±0,01
16	1392	Eugenol	77,53 ±1,38	72,40±3,07
17	1398	β – elemene	0,01 ±0,00	0,01±0,01
18	1410	α – elemene	0,01 ±0,01	0,02±0,01
19	1435	γ-Muurolene	0,02 ±0,02	0,07±0,02
20	1440	γ- cadinene	0,02 ±0,01	0,04±0,02
21	1458	α – farnesene	0,07±0,06	0,13±0,13
22	1469	β- selinene	0,01 ± 0,00	0,02±0,01
23	1469	δ- cadinene	0,13 ±0,16	0,26±0,26
24	1494	β - caryophyllene	5,90 ±1,75	12,61±0,41
25	1507	Caryophyllene epoxide	0,33 ±0,13	0,32±0,06
26	1537	Calamenene	0,06 ±0,01	0,11±0,01
27	1552	Eugenyl Acetate	12,07 ±1,76	9,59±1,61
28	1579	α – humulene	0,84 ±0,23	1,56±0,17
29	1592	Humulene oxide	0,05 ±0,01	0,04±0,01
30	1677	Caryophylla-4(12),8(13)-dien-5.beta.-ol	0,15 ±0,03	0,10±0,01
31	1733	Benzyl benzoate	0,05 ±0,02	0,04±0,01
32	1954	Benzyl salicylate	0,01 ±0,00	0,01±0,00

Based on table 1, it showed the main components in whole clove bud were eugenol, caryophyllene and eugenyl acetate. Whole clove oil contains eugenol (77.53%), β-caryophyllene (5.90%) and eugenyl acetate (12.07%). Crushed clove oil contains eugenol (72.40%), β-caryophyllene (12.61%) and eugenyl acetate (9. 59%).

The same constituents of the oil from whole clove buds and crushed clove buds were methyl heptyl ketone, linalool, benzyl acetate, methyl salicylate, 4-allylphenol, 3,7-dimethylocta-2,6-dien-1-ol,4-allylphenyl acetate, eugenol methyl ether, α-cubebene, β-elemene, α-humulene, alloaromadendrene, γ-muurolene, β-selinene, α-farnesene, γ-cadinene, calamenene, δ-cadinene, caryophyllene epoxide, humulene oxide, caryophylla-4(12),8(13)-dien-5.beta.-ol, benzyl benzoate, and benzyl salicylate.

Other components in whole clove bud were 2-heptanone, 5 metylfuran-2-carbaldehyde 6-methylhept-5-en-2-one, ethyl hexanoate, and benzyl alcohol, whereas the components in crushed clove oil were not identified.

Effect of particle size on the rate of distillation and main constituent of clove oil shown in fig. 1 to fig.3.

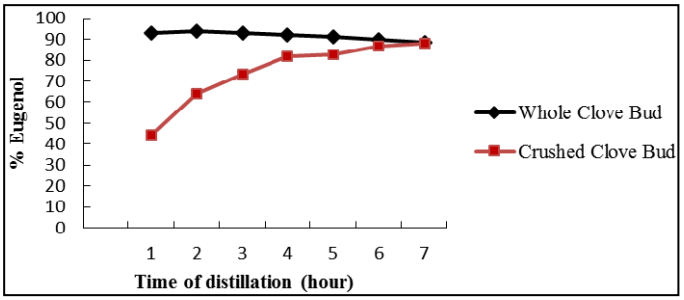

Fig 1: Composition of eugenol vs time of distillation

Based on figure 1, the components of eugenol in whole clove oil was higher than crushed clove oil. The composition of eugenol on crushed clove bud oil increased sharply from one hour to seven hours of distillation. Eugenol in whole clove oil decreased after two hours in the distillation process. Percentage of eugenol on one hour distillation in whole clove bud was 78.16 and crushed clove bud was 44.54%. Time of distillation for seven hours showed the eugenol in whole clove bud was 88.56% and crushed clove bud was 87.85%.

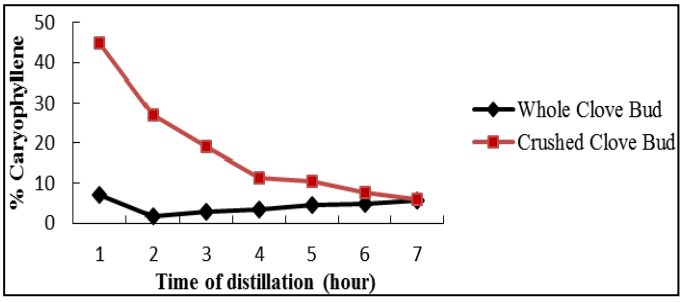

Fig 2: Composition of caryophyllene vs time of distillation

Based on Figure 2, the component of caryophyllene in whole clove bud oil was lower than crushed clove oil. Composition of caryophyllene decreased in whole clove oil and crushed clove oil from one hour to seven hours of distillation. Percentage of caryophyllene on one hour distillation in whole clove bud was 7.25% and crushed clove bud was 44.92%. Time of distillation for seven hours showed the caryophyllene in whole clove bud was 5.17% and crushed clove bud was 12.83%.

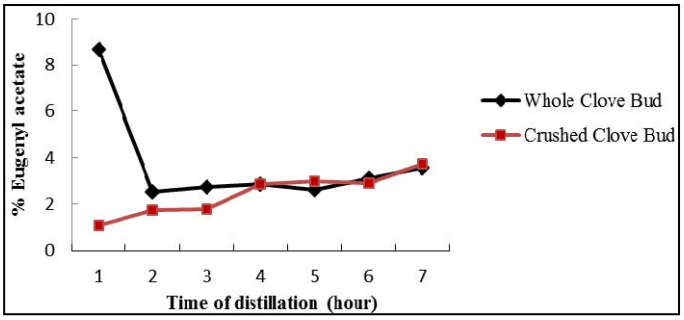

Fig 3: Composition of eugenyl acetate vs time of distillation

Based on Figure 3, the component of eugenyl acetate in whole clove bud oil was higher than crushed cove oil. Distillation at one hour in whole clove bud was 8.69% and crushed clove oil was 1.07%.

4. Conclusion

The particle size of Indonesian clove bud (whole clove bud and crushed clove bud) contributed to distillation rate of essential oil and composition of main constituents of clove oil in particular are eugenol, caryophyllene, and eugenyl acetate. Effect of crushing of clove bud increased caryophyllene, but reduced eugenol and eugenyl acetate in oil compositions significantly.

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