

Physicochemical Properties of Nutmeg Oleoresin Obtained by Ultrasound-Assisted Extraction from Different Raw Material Qualities and Drying Methods

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Abstract— Oleoresin is one of the processed nutmeg products that become the potential agro-industrial export product for Indonesia. The main problems of oleoresin production are the quality diversities of nutmeg as the raw materials because of the improper and inadequate drying process. Ultrasound-assisted extraction (UAE) has recently proposed to shorten the extraction time, increase recovery yield, and also improve oleoresin quality. This study aims to determine the effect of the raw material qualities and drying methods on the yield and physicochemical properties of the nutmeg oleoresin obtained by UAE. Nutmeg was dried with two drying methods (smoked drying and sun-drying) and graded into two kinds of nutmeg quality (grade 1 and 2), then ground into the particle size of 60 mesh. UAE was applied on nutmeg powders with ethanol solvent (1:4 w:v) at 20 kHz frequency, 700 W power, 90% amplitude, at 50°C maximum temperature, and 45 minutes of extraction time. Maceration was carried out as the control treatment by 7 hours of extraction time at room temperature. The raw material qualities and drying methods produced oleoresin obtained by UAE with yield, and physicochemical properties (specific gravity, refractive index, and non-volatile content) were statistically different among treatments. The nutmeg oleoresin obtained by UAE from grade 1 and smoked drying has the highest yield (23.63%) and the lowest non-volatile content (35.54%). The volatile compound as identified by GCMS with 35 total compounds by representing 93.18% of the volatiles where *isocoumarin* (20.30%), *myristic acid* (16.81%), *myristicin* (14.76%), *dehydrodiisoeugenol* (11.11%) dominated.

Keywords— Nutmeg seed; oleoresin; ultrasound; extraction.

Manuscript received 12 Nov. 2018; revised 22 Mar. 2020; accepted 6 Jun. 2020. Date of publication 28 Feb. 2021.
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I. INTRODUCTION

Myristica fragrans Houtt. is a nutmeg plant that becomes a vital plantation crop among various types of nutmeg and other spices. *M. fragrans* is a tropical tree possessing aroma and taste, aboriginal to Banda islands, the Moluccas in Indonesia, and well becomes the largest producer and exporter of nutmeg [1]. Nutmeg is one of the significant commercially products of *M. fragrans* that refers to the dried kernel of the ripe seed. Besides, oleoresin from nutmeg is also commercially used as a flavor, fragrance, and pharmaceutical industries [1], and it can be a potential agro-industrial export product for Indonesia. Oleoresin is a viscous oil from a mixture of essential oils and resins which has characteristics flavor, aroma, and chemical composition resembling the original spices. In contrast to essential oils, oleoresins can resist due to high-temperature processing [2]. Moreover, the oleoresins

contain more volatiles and lipophilic compounds to produce a complete flavor profile and uniformity of the stronger smell, aroma, and taste [3].

The main problem of oleoresin extraction is quality diversities due to the high variation of raw material and imperfect drying process. Both have an impact on the yield, the presence of typical compounds of the plant extract, and its bioactivity [4], [5]. In general, oleoresins are obtained by extraction through conventional methods include distillation, solvent extraction, cold pressing, and sublimation. The method generally runs slowly, decomposition of some volatile compounds, and produces low optimal yield [3], [6].

One of the newest methods that can improve the quality of oleoresin is Ultrasound-Assisted Extraction (UAE). UAE is a simple and efficient technique that can shorten synthesis stages and operating time, and also provides enrichment of flavor compounds on the extract [3]. Several studies have succeeded in obtaining nutmeg oleoresin using an ultrasound-

assisted extraction process. The different used of extraction time, temperature, and distinct amplitude produced yield, physical properties, chemical components, and sensory properties of oleoresins, which also different [2], [6], [7]. Therefore, the objective of this research is to determine the physicochemical properties of nutmeg oleoresin obtained by ultrasound-assisted extraction from different raw material qualities and drying methods.

II. MATERIALS AND METHOD

A. Sample Preparation

The fully matured (6-month old after flowering) dried seeds of nutmeg were handled and processed directly by the farmer in Tanah Rata, Banda, Central Maluku, with standard practices that were usually done and following the Technical Guidelines of Postharvest Handling of Nutmeg in Indonesia by Ministry of Agriculture. The criteria for the quality of nutmeg without shell taken as a raw material following SNI 0006: 2015 nutmeg in the form of quality nutmeg seeds grade 1 (ABCD) and grade II (SS). Two kinds of seed quality that is grade 1 (old nutmeg, glossy and solid shell, smooth within) and grade 2 (less old, sound shrivel, unsmooth within) used as experimental variables. Nutmeg seeds are dried by the method of sun-drying (8-10 hours per day for seven days) and smoking (5-6 hours per day for five days) at a temperature of 35-40°C and 65-75% RH to reach a maximum water content of 10%. In the process, dried nutmeg seeds separated from the shell by pounding and taking the flesh of the seeds. Nutmeg seeds without shells sorted and graded based on quality, size, and wholeness, then packed in separate airtight plastic according to treatment. Nutmeg was shipping in containers from Banda to Bogor during the 10-day delivery time. Quality characteristics include the unit weight of seeds with weighing, water content using the oven method, and fat content using soxhlet analyzed on nutmeg seeds as raw material. Reducing the size is done by grinding and sifting the nutmeg seeds to obtain a size of 60 mesh. The powder is weighed as much as 200 grams as a treatment unit.

B. Oleoresin Extraction

The ultrasound generator (Qsonica, Model 700, 20 kHz, USA) was adjusted the 90% amplitude, 100% output power (700 W) and 45-minute extraction time of the process while the transducer, probe and temperature sensor are dipped in the solution to maintain 50°C maximum temperature applied as the primary treatment. The ultrasound probe with a 25.4 mm diameter replaceable tip immersed in a mixture of approximately 80 mm depth. The extraction was carried out by dissolving 200 grams of nutmeg powder with 800 mL of 96% ethanol in a beaker glass (1:4 w/v powder to solvent ratio). Then the filtrate was filtered and evaporated under vacuum using a rotary vacuum evaporator (B-491 Buchi set) at 50°C until a viscous extract obtained. The various oleoresins then filled in dark glass bottles, subsequently were performed triplicate on four material combinations (grade 1 and sun-drying, grade 2 and sun-drying, grade 1 and smoked drying, grade 2, and smoked drying). The mean value of the yield, specific gravity, and color was measured. The combination of grade 1 nutmeg seeds and smoked drying was extracted for 7-hour at room temperature (30°C) by the

maceration technique as the control treatment. Every 10 minutes during the extraction time, the solution was manually stirring.

The temperature set to a maximum of 50°C through the control that takes place during the extraction process. The beaker placed in a container filled with water as a cooling medium. The temperature sensor is placed in a beaker to record temperature fluctuations in the computer (recorder). The temperature of the solution in the beaker increases the temperature of water in the container also rises. When the extraction temperature reaches the maximum temperature, the water pump turns on to replace the water in the container into cold water, thereby reducing the temperature of the extraction solution—the circulation of water in the container done through a hose equipped with copper pipes in the icebox. The copper pipe lowers the temperature of the water and returns to the container to maintain the temperature during extraction. Then the extraction results are filtered, the filtrate evaporated with a rotary vacuum evaporator at a pressure of 200 mbar and a temperature of 50°C. The evaporation process stopped if air bubbles formed, which continuously break on the surface of the thick extract, and the solvent stops dripping on the flask to produce oleoresin. The method scheme of ultrasound-assisted extraction as shown in Fig. 1.

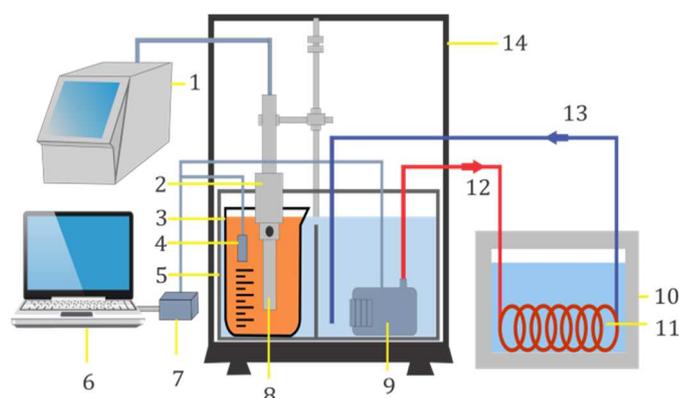


Fig. 1 Ultrasound-assisted extraction method scheme (1. ultrasonic generator; 2. transducer; 3. beaker glass; 4. temperature sensor; 5. Water bath; 6. recorder; 7. microcontroller; 8. probe; 9. dip pump; 10. icebox; 11. copper pipe; 12. hot water flow; 13. cold water flow; 14. sonicator room)

C. Physicochemical Analysis

Oleoresin components extract were analyzed to obtain data in the form of yield with weighing and physicochemical properties. The refractive index analyzed using the ABBE refractometer (Otago, USA). The specific gravity using a pycnometer. Non-volatile content through the evaporation process in water baths, and solubility in ethanol 90% based on SNI 06-2388-2006 procedures concerning nutmeg oil. Chemical components were analyzed using Shimadzu GC MS-QP 2010S Ultra gas chromatograph (Shimadzu Corporation, Kyoto, Japan). GCMS equipped with Capillary Column Model Number: Agilent 19091S-433 HP 5MS 5% Phenyl Methyl Siloxane (inner diameter 250 μm , length 30 m, and film thickness RTX-5 column of 0.25 μm) and FID as a flame ionization detector used. The initial temperature of GC condition at 60°C then raised to 250°C (4°C/min), while the column was maintained 250°C for 20 minutes with a flow rate of helium carrier gas set at 20 mL/min. Compound identification made by comparing the retention index and

mass spectra through the database. Compounds are grouped based on the similarity of each compound $\geq 70\%$. The number of compounds is expressed in percent (%) area by computerized matching with Wiley Library.

D. Statistical Analysis

Triplicate samples of nutmeg oleoresins for each treatment and control were taken, and each sample was analyzed individually in triplicate. ANOVA analyzed the data with Randomized Factorial Design for UAE treatment and Duncan's multiple range test used for further tests. The quantitative data of UAE and maceration statistically examined by paired T-test. The probability value of $P \leq 0.05$ used to denote significant differences. Research data were processed using SPSS.

III. RESULT AND DISCUSSION

A. The Quality of Raw Materials and Yield of Nutmeg Oleoresins

The size of Banda nutmeg characterized with weight of 4.2834 - 5.7846 g/grain as dried seed on this research. Besides, other research confirmed that Banda Sangihe nutmeg has seed weight 6.9 - 9.5 g/grain. The size and nutmeg yield found high variation among plantations in Indonesia related to varieties, seasonal weather, and cultivation practices [8]. However, the nutmeg seeds in the research have perfectly shaped, relatively heavy, not attacked by pests and diseases, and did not get mechanical damage.

Further test results in Table 1 show that the grade 1 nutmeg seeds by smoked drying have the highest seed weight (5.7846 g) and highest fat content (35.27%). The combination of these treatments, based on the results of further tests in Table 2, produced the highest yield (23.63%). On the other hand, the combination of grade 2 nutmeg seeds by sun-drying has the highest water content (10.51%) and produced the lowest yield (13.42%). The age of harvesting affects the quality of raw materials, grade 1 nutmeg seeds come from fruits that picked quite old while grade 2 nutmeg seeds come from less old seeds. Early harvesting can cause shriveled, cracked, and broken kernels easily in the drying process and storage duration [9].

TABLE I
CHARACTERISTIC OF BANDA'S NUTMEG AS RAW MATERIAL

Physical properties	Sun-drying		Smoked drying	
	Grade 1	Grade 2	Grade 1	Grade 2
Weight (g) ^c	5.7229 ± 0.1649 ^a	4.2834 ± 0.1032 ^c	5.7846 ± 0.1348 ^a	4.5317 ± 0.1300 ^b
Moisture content (%db) ^f	7.92 ± 0.04 ^c	10.51 ± 0.08 ^a	7.81 ± 0.01 ^d	10.21 ± 0.07 ^b
Fat content (%db) ^f	33.45 ± 0.02 ^b	24.89 ± 0.01 ^d	35.27 ± 0.03 ^a	28.80 ± 0.01 ^c

^{a, b, c, d} Numbers with different letters in each characteristic show a significantly different effect ($\alpha = 0.05$) based on Duncan's multiple range test.

^e Mean value of 10 replications from each whole nutmeg

^f Mean value of 3 replications from each nutmeg powder

The drying temperature and time exposure also become the parameter that could affect the moisture contents of dried nutmeg. The higher the temperature, the lower the humidity,

then the difference of water vapor pressure inside the nutmeg with the outside can increase water removal and the rate of drying. Moreover, the high temperature can cause nutmeg to become shriveled. Besides, the drying rate is influenced by the shape, size, and initial moisture content of nutmeg [9], [10]. However, the higher the temperature the higher the risk of complete loss of moisture due to prolonged exposure [11].

Seed weight, water content, and fat content are related to harvest age. The older the harvest, the seed weight, fat, and oil content increases. The hard-old nutmeg seeds increase the percentage of nutmeg particles that pass the sieve to produce a high yield. Young nutmeg seeds have high water content and soft texture so that the surface of the seeds shrink after drying [11]. The heat transfer and mass transfer during drying will depend on surface area increases. A higher surface area leads to the maximum escape of water content. High water content in young nutmeg affects the speed of oil diffusion, thereby reducing the yield due to the dry outer layer and water molecules that inhibit oil extraction [5]. Moreover, the higher water content will increase the growth of microorganisms that could spoil the quality of oleoresin [6], [10].

Nutmeg seeds dried in controlled dryers such as mechanical dryers, ovens, and curing have a constant temperature and heat source resulting in higher oil content, fat content, and oleoresin yield than sun-drying [4]. Sun-drying method is kind of no control temperature drying which leads to improper method that causes the risk of compound loss such as myristicin oil due to high temperature during the exposure time [9], [10]. Other studies deduced that drying methods as the main effect of extract concentration on *Vernonia amygdalina* leaf [5]. The type of drying which gives varying on drying temperature also affected to the yield and bioactivities of polysaccharides due to chemical composition and molecular structure changing in *Symphytum officinale* L. [12]. The different drying method which gives the different temperature and drying time also affected to the optimum quality of ginger and red ginger simplicial [10].

The evaporation of volatile content decreases the yields during drying [4]. Oil content related to water content, the higher the water content the smaller the oil yield obtained. Materials that initially contained much water in cells undergo diffusion while bringing volatile compounds to the surface until evaporation occurs. Oil evaporation also will occur along with water vapor during the drying process, so the longer the material is dried the more oil is evaporated. In the process, sun-drying requires a longer drying duration than smoked drying. More extended drying periods cause differences in the percentage of essential oils and fatty oils, due to evaporation on the effect of potentially higher temperatures [9].

Statistical analysis showed that ultrasound-assisted extraction (UAE) resulted in higher yields of nutmeg oleoresin compared to maceration (Table 2). The amplitude used in ultrasound-assisted extraction in this study is a quantity that has almost reached its maximum, thus producing sizeable total energy due to cavitation. Enormous total energy increases the temperature to be higher, thus increasing the diffusion of solvents in a substance due to the presence of ultrasound bubbles and cavitation intensity. Ultrasound waves create bubbles in the solvent that break near the cellular structure and produce a high-speed solvent stream. The

solvent stream can increase the penetration and diffusion of solvents into cells, cause enlargement of cell walls, and accelerate overall mass transfer rate [2].

The higher the temperatures of ultrasound-assisted extraction, the higher the yields of nutmeg oleoresin. The higher temperature makes the viscosity of the solvent decrease so that the extraction mechanism and the rate of oleoresin transfer from the material to the solvent are faster. An increase in temperature due to the cavitation bubble collapse makes the pores of the particles expand and facilitates the solvent to diffuse easily [3]. Contact between the solvent and the material increases so that the solvent is

easier to penetrate the cell even in a short time due to a violent shock wave and a high-speed jet were produced [13].

B. Specific Gravity and Refractive Index of Oleoresins

The combination of raw material quality and drying method produced specific gravity and refractive index of oleoresin from ultrasound-assisted extraction with significantly different (Table 2). The specific gravity of nutmeg oleoresin obtained by UAE is higher than the standard. SNI 06-2388-2006 stated that the specific gravity of nutmeg is in the range of 0.880-0.910. Besides that, the refractive index of nutmeg oleoresin obtained by UAE is following the standard, which is in the range of 1.470-1.497.

TABLE II
OBSERVATION DATA OF PHYSICAL PROPERTIES OF NUTMEG OLEORESIN OBTAINED FROM ULTRASOUND-ASSISTED EXTRACTION AND MACERATION TECHNIQUE

Physical properties	Maceration	Sun-drying		Smoked drying	
		Grade 1	Grade 2	Grade 1	Grade 2
Yield (%db)	12.30 ± 0.00	21.70 ± 0.00 ^b	13.42 ± 0.00 ^d	23.63 ± 0.00 ^a	17.69 ± 0.00 ^c
Specific gravity (20/20°C)	1.1330 ± 0.0030	1.0389 ± 0.0052 ^{ab}	1.0160 ± 0.0033 ^a	1.0685 ± 0.0227 ^c	1.0404 ± 0.0072 ^b
Refractive index (20°C)	1.4706 ± 0.0010	1.4898 ± 0.0035 ^{ab}	1.4945 ± 0.0009 ^a	1.4921 ± 0.0045 ^a	1.4863 ± 0.0032 ^b
Nonvolatile content (%db)	43.27 ± 0.06	36.69 ± 0.01 ^c	40.88 ± 0.08 ^a	35.54 ± 0.10 ^d	38.57 ± 0.01 ^b
L	22.86 ± 0.05	25.09 ± 0.15 ^c	23.77 ± 0.17 ^b	23.81 ± 0.11 ^b	23.06 ± 0.19 ^a
a	2.02 ± 0.02	4.35 ± 0.03 ^c	0.75 ± 0.01 ^a	4.93 ± 0.51 ^d	1.30 ± 0.01 ^b
b	0.91 ± 0.01	2.34 ± 0.01 ^c	0.45 ± 0.00 ^a	3.17 ± 0.01 ^d	0.83 ± 0.01 ^b
C	2.21 ± 0.02	4.94 ± 0.02 ^c	0.87 ± 0.01 ^a	5.86 ± 0.02 ^d	1.54 ± 0.01 ^b
H	24.24 ± 0.21	28.22 ± 0.25 ^a	31.06 ± 0.27 ^b	32.74 ± 0.10 ^c	32.50 ± 0.19 ^c
Color			Red		
90% Ethanol solubility			1:1 soluble, distinct opalescence		

^{a, ab, b, c, d} The mean value of triplicate, numbers with different letters in each physical property show a significantly different effect ($\alpha = 0.05$) based on Duncan's multiple range test.

The specific gravity and refractive index of oleoresin produced from grade 1 nutmeg seeds is higher than grade 2, except the refractive index of nutmeg seeds resulted from sun-drying. The higher the quality of raw materials, the higher the value of specific gravity and refractive index of oleoresin. So that the older the harvest age, the higher the specific gravity and oleoresin refractive index. That is because the earlier the time of harvest, the lower the formation of secondary metabolites in line with the high production of primary metabolites. Besides, the earlier the time of harvest, the higher the weight fraction in the composition of the compound compared to the lighter fraction [14].

The specific gravity and refractive index of oleoresin produced from smoked drying nutmeg seeds is higher than sun-drying. Sun-drying requires a longer drying duration so that it has a higher temperature effect than smoked drying. The statistical analysis showed that nutmeg oleoresin from maceration has the highest specific gravity and the lowest refractive index compared with the ultrasound-assisted extraction oleoresin (Table 2). The differences in specific gravity can be affected by temperature and duration of heating during the drying, extraction, and evaporation processes. Oleoresin obtained from the production process with the use of high temperatures has higher specific gravity than the standard. The higher specific gravity caused by the formation of resin concentrates. Volatile compounds continue to evaporate during the heating process so that more weight fractions with the highest boiling point of other compounds of nutmeg are remained [15].

C. Color of Nutmeg Oleoresins

Nutmeg oleoresin has a red color. Further test results in Table 2 show that nutmeg oleoresin has significantly different L, a, b, chroma, and hue between treatment combinations. The combination of grade 1 nutmeg seeds and sun-drying has the highest L value, while the combination of grade 1 nutmeg seeds and smoked drying has the highest values of a, b, chroma, and hue.

Drying affects the color of oleoresin because of the potential for color changes in the raw material [11]. Mechanical drying, oven, and smoked drying produce a better color of raw materials and oleoresin than sun-drying [4]. Besides, the quality of raw materials affects the color of oleoresin due to differences in harvest age [14]. The older the harvest age, the color of the nutmeg shells getting brown to black and mace become bright reddening [1]. Generally, nutmeg oleoresin reported having a red, yellow, and red-brown color on the filtrate and as a thick liquid after the evaporation process [6], [7].

D. The Solubility of Nutmeg Oleoresins in 90% Ethanol

The solubility of nutmeg oleoresin in 90% ethanol showed the same results between the ultrasound-assisted extraction and maceration (Table 2). The quality of the raw materials, the drying methods, and the extraction methods did not affect the solubility of nutmeg oleoresins in 90% ethanol. The solubility of nutmeg oleoresin in 90% of ethanol is 1:1 (oleoresin:solvent v:v). This result is following the standards

of SNI 06-2388-2006 concerning nutmeg oil with a maximum value of 1:3 (*oleoresin:solvent v:v*) [15].

The solubility of oleoresin in ethanol is related to polarity. The higher the need for ethanol to dissolve, the more difficult oleoresin to dissolve. The solubility of oleoresin in 90% ethanol used to evaluate the resignification of essential oil. If many compounds containing OH groups in volatile contained, the higher the solubility of oleoresin. Conversely, if most terpene compounds contained in oleoresin, then the solubility decreases and generates competition among hydrophobic compounds. However, the difference compound that affects the solubility will vary due to the loss by volatilization and leaving the resin [16]. Generally, nutmeg oleoresin has a high volatile compound such as myristicin which is insoluble in water but soluble in organic solvents [16]. Therefore, the mechanism of resignification that occurs is still following the standard of solubility.

E. Nonvolatile Content and Chemical Composition of Nutmeg Oleoresins

The results of further tests in Table 3 show that the oleoresin from the combination of grade 1 nutmeg seeds and smoked drying obtained by UAE has the lowest nonvolatile content (35.54%). Statistical analysis also showed that maceration produced nutmeg oleoresin with the highest nonvolatile content (43.27%) compared to UAE. The remaining evaporation represents the content of nonvolatile

compounds which are dominated by fat oil (fixed oil) or other compounds with high molecular weight [15][16].

Based on the data in Table 2, it stated that the nutmeg oleoresin obtained by UAE contains 35.54-40.88% fatty oil with 59.12-64.46% essential oil component (7.93-15.23% of the total materials). This value is different from macerated nutmeg oleoresin containing fat oil by 43.27%, with the essential oil component of 56.73% (6.97% of the total materials). Nutmeg seeds contain essential oils and fatty oils, depending on the origin of the raw material. Other study revealed the result of 3.11-3.91% essential oils and 14-15% oleoresin of nutmeg seeds from Papua [8]. Generally, the essential oil content of nutmeg seeds from Papua is much lower than the essential oils of Banda nutmeg seeds.

The combination of raw material quality and drying method produced oleoresin by UAE with the same main compound profiles, namely *myristic acid*, *isocoumarin*, *myristicin*, and *dehydrodiisoeugenol*. Hence the grade 1 nutmeg seeds with sun-drying produced the profile of the main UAE compounds, namely myristic acid, glycerin, and myristicin. In contrast to the profile of the main compounds resulting from maceration, namely *methoxyeugenol*, *myristicin*, and *dehydrodiisoeugenol*. Nutmeg seeds grade 2, which are dried by smoking, produced the highest total components (38 components). While nutmeg seeds grade 2 dried by sun-drying produced the highest total constituents (94.93%) (Table 3).

TABLE III
CHEMICAL COMPOSITION PROFILE OF BANDA'S NUTMEG OLEORESIN OBTAINED BY ULTRASOUND-ASSISTED EXTRACTION AND MACERATION TECHNIQUE

Chemical Compounds	Retention Time (RT)	Type*	The yield of oleoresin constituents (%)				
			Maceration	Sun-drying		Smoked drying	
				Grade 1	Grade 2	Grade 1	Grade 2
<i>Glycerin</i>	2.300	3	0.64	16.46	0.18	0.50	0.71
<i>Isoeugenol</i>	6.994	1	2.14	1.24	2.09	1.96	2.11
<i>Myristicin</i>	7.866	1	19.84	11.02	10.57	14.76	13.28
<i>Methoxyeugenol</i>	8.729	1	26.08	9.07	8.04	6.72	7.82
<i>Myristic acid</i>	10.354	2	1.66	32.54	24.70	16.81	6.13
<i>Palmitic acid</i>	12.072	2	0.85	3.59	3.03	2.25	1.56
<i>Oleic acid</i>	14.149	2	0.46	2.43	2.49	1.70	1.94
<i>Dehydrodiisoeugenol</i>	17.945	1	11.44	6.17	10.87	11.11	11.57
<i>Acetophenone</i>	18.193	1	2.19	-	1.78	1.89	2.34
<i>Isocoumarin</i>	18.458	1	2.23	-	18.14	20.30	22.97
<i>26,27-Dinorcholest-5-en-23-yn-3. beta. -ol</i>	20.526	4	2.04	1.18	2.02	2.04	2.49
<i>5-Isopropenyloxyethylene-3,3-dimethyl-cyclohexanone</i>	20.894	5	5.84	-	4.50	3.70	6.44
Others ^a			8.90	7.21	6.52	9.44	9.62
*Type							
1. <i>Aromatic acids</i>			67.67	27.50	54.39	60.76	64.78
2. <i>Carboxylic acids</i>			3.61	40.35	30.75	20.85	9.73
3. <i>Polyols</i>			0.64	16.46	0.18	0.5	0.71
4. <i>Sterols</i>			2.04	1.18	2.02	2.04	2.49
5. <i>Enols</i>			5.84	-	4.50	3.70	6.44
6. <i>Miscellaneous</i>			4.51	5.42	3.09	3.53	4.83
Total ^b			84.31	90.91	94.93	93.18	88.98
Total identified compounds			32	12	31	35	38

^a Identified components by $\leq 2\%$ area and less than two kinds of oleoresins are not showed

^b Identified components by $\leq 70\%$ similarity excluded

Myristicin is a safrole derivative with a methoxy group which determines the quality because myristicin gives a distinctive aroma to nutmeg oleoresin [15]. Myristicin acts as an anticancer by inhibiting cancer cell apoptosis in human neuroblastoma [1]. While myristic acid is long-chain saturated fatty acid with 14 carbon chains obtained from the

trimyristin saponification process. Also, desaturation process of myristic acid (C14) to *tetradecadienoic acid* in plants by fatty-acyl pathway derived the pheromone components for beetle [17]. Myristic acid, together with palmitic acid, can be use as the core phase change material for thermal energy

storage by giving the thermal stability properties through microencapsulation process [18].

Dehydrodiisoeugenol (DDIE) is a bioactive neolignane that functions as a model for the structural units of *biphenyl* and *phenylcoumaran* in the lignin molecule. DDIE provides good anti-inflammatory and anti-oxidative activity. This compound revealed to pass the cytotoxicity test and has the same strong effect as commercial antioxidants such as BHA and BHT [1].

Also, *methoxyeugenol* which becomes the main compound in macerated nutmeg oleoresin is called O-dimethyl elemicin, this compound is a metabolite formed from elemicin. In contrast to psychedelic allylbenzenes groups such as elemicin and myristicin, *methoxyeugenol* contains a hydroxy group at position 3-benzene. This group is a substrate of glucuronosyltransferase (UGT) and sulfotransferase (SULT) which can reduce the lipophilic properties of *methoxyeugenol*. *Methoxyeugenol* is a natural antibacterial that can inactivate *Escherichia coli* bacteria in the application of pulsed electric field technology [19].

The combination of raw material quality and drying method produced oleoresin with the primary component type in the form of the aromatic group, except oleoresin from grade 1 nutmeg seeds with sun-drying (Table 3). Smoked drying gave the oleoresin by UAE has a higher aromatic content (60.76 - 64.78%) than sun-drying (27.50 - 54.39%), but both are lower than macerated oleoresin (67.67%). On the contrary, sun-drying gave the oleoresin by UAE has a higher carboxylic acid content (30.75 - 40.35%) than smoked drying (9.73 - 20.85%) and macerated oleoresin (3.61%). Nutmeg oleoresin of grade 2 (74.51 - 85.14%) has a significant component type in the form of aromatic and carboxylic acids, which are higher than grade 1 (67.85 - 81.61%) in both drying methods.

The drying method applied to the raw material will affect the content of oleoresin, especially essential oils [4]. The volatile component is reduced during drying at temperatures higher than room temperature due to diffusion with water until evaporation occurs. The drying period also causes differences in the percentage of essential oils and fatty oils, due to evaporation on the effect of potentially higher temperatures. The higher the temperature and duration of drying make the volatile compounds sensitive to oxidation gradually decrease with the effects of heat [9]. The drying process can cause uneven heating and destructive effects on the heat-sensitive compound of nutmeg [3]. While higher quality results in lower non-volatile content, it is related to the age of nutmeg harvest. The older the harvest age, the content of secondary metabolites, mainly aromatic and carboxylic acid contents, increased with decreasing content of terpenes. The content of secondary metabolites in the form of carboxylic acid groups and the cyclization of aromatic acid groups is a weight fraction that has the highest molecular weight compared to other groups of compounds [14].

The heating process can raise new compounds and removes other compounds. This happens because the compound has reached the boiling point so that the chemical composition is different [15], [16]. Some compounds can be extracted by ultrasonic-assisted extractions, such as the aromatic group, which cannot be identified in maceration-induced nutmeg oleoresin. Ultrasonic-assisted extractions provide a higher intensity of flavor composition than maceration as the

conventional method. Ultrasound provides energy to break oil glands to increase the release of essential oils [3]. Besides, the use of higher ultrasonic intensity can extract high carboxylic acid compounds [2].

IV. CONCLUSION

It is concluded that physicochemical properties (specific gravity, refractive index, and nonvolatile content) are directly proportional to raw material quality; whereas smoked drying gives higher value than sun-drying. The combination of grade 1 nutmeg seeds and smoked drying yielded the highest yield of ultrasound-assisted nutmeg oleoresin (23.63%) and the lowest nonvolatile content (35.54%) compared to other treatment combinations. Maceration produced nutmeg oleoresin with the lowest yield (12.30%) and the highest nonvolatile content (43.27%) compared to UAE. GCMS identified 12 – 38 total compounds by representing 88.98 – 94.93% of the volatiles varied at the different raw material quality and drying methods. The main chemical compounds of nutmeg oleoresin were *myristic acid*, *isocoumarin*, *glycerin*, *myristicin*, *dehydrodiisoeugenol* and *methoxyeugenol* dominated as many as 47.82 – 64.28% with different composition on each treatment combination. The ultrasonic extraction has given the physicochemical properties of oleoresin at 20 kHz frequency, 700 W power, 90% amplitude, 50°C maximum temperature and 45-minute better than 7-hour and 30°C temperature maceration technique. Ultrasound-assisted extraction can be a promising approach to extract the oleoresin in a shorter time which gives the maximum quality and quantity.

ACKNOWLEDGMENT

The authors are grateful and acknowledge for the research funding from Directorate of Research and Community Service, Ministry of Research Technology and Higher Education of the Republic of Indonesia through the National Strategic Research Program (PSN) with Contract Number of Agreement: 129 / SP2H / PTNBH / DRPM / 2018.

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