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Ultrasound Assisted Extraction of Oleoresin from Nutmeg (Myristia Fragrans Houtt)

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Abstract— **In the present study, the application of ultrasound to extract oleoresin from nutmeg (Myristica fragrans Houtt) was investigated. The extraction of oleoresin from nutmeg meat was performed by using ethanol as solvent. Experiments were carried out under the following conditions: total weight of nutmeg meat of 200 g, the mass ratio of dry meat nutmeg to solvent of 1:4, the size of the material of -10+18 mesh and the ultrasonic frequency of 45 kHz; with variation of extraction temperature (30, 40 and 50°C), and extraction time (1, 2 and 3 hours). This research was also using maceration and soxhlet for comparison. The experimental results showed that the Soxhlet method requires a longer time and higher temperature to produce oleoresin compared with maceration and ultrasonic waves assisted extraction methods. The ultrasonic extraction resulted highest oleoresin yield of 6.2% was obtained at the condition of 50oC and 3 hours of extraction temperature and extraction time, respectively. The oleoresin produced in the experiment was analyzed by gas chromatography-mass spectrometry (GC-MS), which showed that the two largest components in oleoresin are myristicin and eugenol.**

***Keywords*— Nutmeg; Oleoresin; Extraction; Ultrasound.**

I. INTRODUCTION

Nutmeg has been used long enough, either in traditional or conventional. These efforts include nutmeg oil processing which currently has a high selling price. The nutmeg plant needs to be managed, developed, and utilized optimally with the rapid growth. Meat nutmeg is the largest portion of the nutmeg and can be processed into oleoresin.

Oleoresin of spices are used in industrial scale, is commonly used to flavour the food processing industry-like meat canning, sauces, soft drink manufacturing, pharmaceutical raw materials, cosmetic and perfume industry, confectionery and bakery industries [1].

The need for effective extraction of biologically active component from plants without any loss of activity and high purity has resulted in developments of newer processes of extraction. Various mechanical and chemical processes such as solvent extraction [2], steam distillation [3], high hydrostatic pressure extraction [4], pulse electric field process [5], high pressure process [6], etc are used for the extraction of product from plant materials. The selection of the method to extract active components with maximum yield and highest purity mainly depends on the nature of compound and thermal stability and nature of raw material to be processed. It is well known that conventional extraction techniques are often limited by the mass transfer resistances due to involvement of more than one phase in the system [7].

Cavitation generated using ultrasound is known to produce physical effects such as liquid circulation currents and turbulence with can lead to a significant increase in the mass transfer rates. Ultrasound can be effectively used to increase the yield and rate of mass transfer in several solid liquid extraction processes. The mechanism of intensification of extraction process has been attributed to cavitation phenomena resulting in intense turbulence and liquid circulation currents in the system [7-10]. The difference substances that have been extracted using ultrasound include essential oils, aromatic compounds, citrus compounds, sugar, proteins, acids, natural dyes, pigments, etc. In addition to beneficial effect of ultrasound on the yield and kinetics of extraction, it has been observed that extraction can be carried out at much lower temperature and pressure which can result in substantial decrease in the overall cost of the operation. It is however important that the sonication time has to be carefully optimized, since exposure to ultrasonic irradiation may damage the quality of the solute in some cases of heat sensitive materials, etc. [11]

Ultrasonic waves are longitudinal mechanical waves with frequencies above 20 KHz which is the area limit of human hearing. Ultrasonic waves can propagate in a solid medium, liquid and gas. This is caused because an ultrasonic wave propagation of mechanical energy and momentum, the energy propagation depends on the molecular interaction and inertial properties of the medium in its path [12].

Ultrasound can be used to increase conversions and improve the selectivity, so that yield can obtained more. With the use of ultrasonic waves allows the process carried out at pressures and lower temperatures, reducing the use of raw materials and a solvent, reducing the synthesis phase and increase the final selectivity, allowing the use of raw materials and solvents with low purity [13]

The objectives of this research are to increase the economic value of nutmeg by comparing the yield of the maceration method, soxhletation, and ultrasonic assisted extraction to obtain the optimum operating condition with the quality and quantity maximum of oleoresin.

II. MATERIALS AND METHODS

Materials used in this research were the meat of nutmeg (Myristica fargrans Houtt) and ethanol PA (concentration 99%). The equipment used in this research were ultrasonic cleaner (Bransonic 8510), rotary vacuum evaporator (Eyela N-1001 series), soxhlet tool, analytical balance, hot plate, water pump, vacuum pump, boiling flask 500 ml, condenser and several tools to support the research.

The research was divided into 5 steps, i.e. preparation of raw materials, raw material analysis, extraction (soxhletasi, maceration, and ultrasound), oleoresin purification, and product analysis. The production of oleoresin was started from nutmeg meat sorted and cleaned of dirt, washed, drained and dried in the sun for days or in the oven dryer until the water content below 10%. Then, samples was crushed with a ball mill and sieved with a sieve vibrator to produce material size of -10 + 18 mesh. Furthermore, the meat of nutmeg as a raw material was analysed for water content.

After analysing the water content, the sample as much as 200 grams was put into a flask and then added ethanol solvent with the mass ratio of dry meat nutmeg to solvent of 1:4. The ultrasonic frequency was of 45 kHz and the temperature was set according to experimental variables (30, 40, and 50oC) . Then, the variation time of extraction was for 1, 2, and 3 hours. After reaching the extraction time, the filtrate was filtered from the waste and then evaporated from water. Oleoresin product was analysed, in terms of yield, specific gravity, refractive index, and oleoresin components. Figure 1 showed the flow diagram of

oleoresin formation.An easy way to comply with the conference paper formatting requirements is to use this document as a template and simply type your text into it.

III. RESULTS AND DISCUSSION

A. Analysis of Moisture Content in NutmegPowder Document

This research used fresh nutmeg meat that has been separated from other parts of the fruit and then dried and crushed into dry nutmeg powder. The size of the material used in this research was -10 +18 mesh with moisture content was still contained in dry nutmeg powder about 8.2% based on analysis using flour moisture meter. High water in the nutmeg powder greatly affected the condition of the materials to produce high quality of oleoresin. If the moisture content of nutmeg meat contained more than 10%, the possibility of the growth of microorganisms in powdered nutmeg oleoresin will spoil the quality.

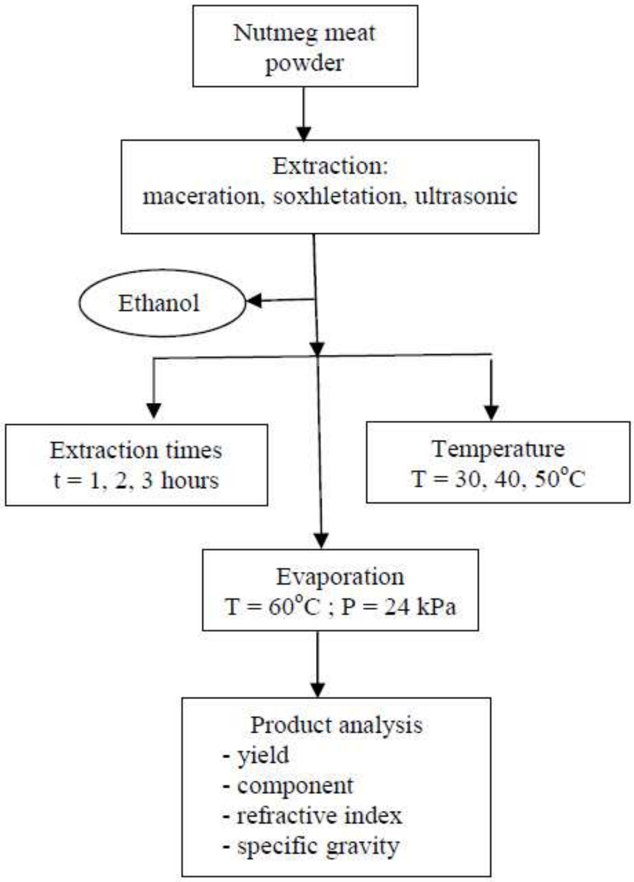

Fig. 1. Flow diagram of oleoresin extraction

B. Product Analysis

1) Effect of Time and Temperature on Oleoresin Extraction: The results of the extraction are shown by the redbrown colour of filtrate. After separating by evaporation, red-brown viscous liquid was obtained. This suggested that the presence of the oleoresin extracted. Extraction time and temperature affected the amount of the resulting yield of oleoresin. Based on the results by using extraction with the aid ultrasonic method, it produced the highest yield at 50oC operating conditions with extraction time for 3 hours namely 6.2%. Comparison of yield at each time and temperature was shown in Figure 2.

At the temperatures of 30oC, 40oC, and 50oC for 1 hour obtained each yield were 0.65%, 1%, and 2.3% respectively. It showed that the higher yield when increasing extraction temperature. Increase the temperatures, the pores of the particles will expand and facilitate solvent diffuse into solids. Low temperatures with shorten extraction time will reduce solvent contact with the material so that the solvent will be difficult to penetrate to the cells material. Temperature and extraction time affected oleoresin yield.

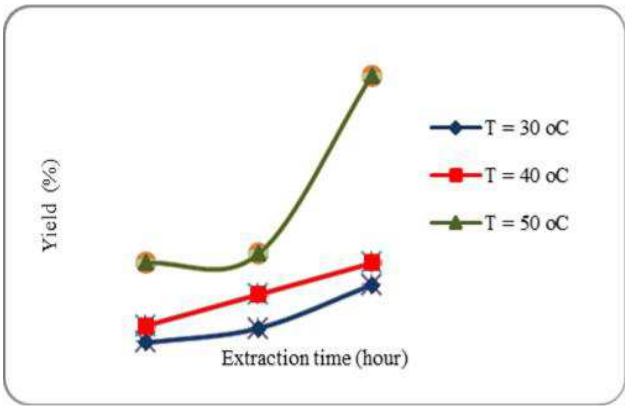

Fig. 2 Relationship oleoresin yield (%) of the extraction time and temperature on the ultrasonic extraction method

Yield increased with increasing of the extraction time at which the extraction temperature 50°C for 1, 2, and 3 hours respectively obtained yield of 2.3%, 2.5%, and 6.2%. According to Jayanudin [14], a longer extraction time provide a greater yield due to the increased solubility of the solvent. It will facilitate solvent into the pores of the solid ground nutmeg and dissolve the components contained in the nutmeg.

2) Effect of Extraction Time and Temperature on Refractive Index and Density of Oleoresin: The refractive index will increase with increasing of temperature and extraction time. Oleoresin has a refractive index smaller than the refractive index of the FAO standard (1.4720 to 1.4860). It can be caused by the thickness of oleoresin. If the oleoresin too thick, it will cause the falling value of refractive index of oleoresin. It can be happened because of the impurities in the nutmeg. [15]. Refractive index of oleoresin is the ratio between air velocity in a vacuum to the velocity of light in oleoresin [15]. From the definition, it can be concluded that the more concentrated oleoresin the lower refractive index and conversely the oloeresin dilute the higher the refractive index value.

TABLE I
OBSERVATION DATA OF SPECIFIC GRAVITY AND REFRACTIVE INDEX FOR OLEORESIN WITH ULTRASONIC EXTRACTION

Time (hour)	Parameters					
	Refractive index			Density (gr/ml)		
	30 °C	40 °C	50 °C	30 °C	40 °C	50 °C
1	1.4375	1.4390	1.4415	0.453	0.522	1.089
2	1.4380	1.4420	1.4395	0.638	1.417	1.050
3	1.4410	1.4385	1.4400	1.250	0.817	1.200

From Table 1, it also showed that the increase of density when increasing extraction time and temperature. Density is the number of molecules per unit area or volume or length per unit volume is usually calculated in g/cm3 or number g /ml. From the results, it can be showed that the condition of the extraction affected the oleoresin density. Extraction process of oleoresin at high temperatures produced oleoresin with high density. This phenomenon was caused by temperature light fraction (volatile substances) disappearances. If the process with high temperature was run, it will be easy to formed more resin as a non volatile compounds [16].

Most oleoresin produced at a temperature of 50oC have a specific gravity higher than the FAO standards, while the density was lower at 30oC and 40oC ranged from 0.880 to 0.910. Density values approaching FAO standard (0.880 to 0.910) was achieved at temperature 40oC with oleoresin extraction time for 3 hours. It was equal to 0.817 gr/ ml. Density can be affected by the length of heating time during evaporation. Long time of evaporation will produce very concentrated oleoresin and it will lead to a high density. It also caused by the evaporation of the high temperature light fraction (volatile substances) of oleoresin, and it will left only a fraction of the weight.

3) Comparison of Ultrasonic Extraction Method with Soxhlet and Maceration Method: Comparison of the extraction methods aimed to review the differences oleoresin produced in terms of quantity and quality. Maceration extraction method was set at the same temperature to the lowest temperature on the extraction with ultrasonic waves at 30oC for 1, 2, and 3 hours. Based on Table 2, it can be seen that the highest yield obtained at soxhlet method. It was equal to 15.3%, but soxhlet method took quite a long time and it was carried out at the boiling point of ethanol for the extraction process. Yield of the extraction was slightly higher than maceration extraction with ultrasonic waves.

By using the maceration and extraction methods with ultrasonic waves, the extraction can be performed at a lower temperature conditions with a shorten time. Refractive index and density also showed the results of the ultrasonic method 30oC for 3 hours the closest standard. By comparing the results of oleoresin from the three methods, it was found that the most favorable conditions was the method of extraction with the aid of ultrasonic waves.

TABLE II
OBSERVATION DATA FOR ULTRASONIC, MACERATION, AND SOXHLET METHOD.

Method	Parameter	Time (hour)		
		1	2	3
Ultrasonic*)	%	0.65	0.95	1.85
	ρ	0.453	0.638	1,.50
	nD	1.438	1.438	1.441
Maceration*)	%	0.8	1.1	2.05
	ρ	0.829	1.015	1.126
	nD	1.438	1.439	1.440
Soxhlet**)	%	15.3		
	ρ	1.100		
	nD	1.38		

information:
*) = temperature 30oC
**) = 7 hours
% = yield
ρ = density (gr/ml)
nD = refractive index

4) Effect of Operating Conditions on the Extraction Process Components Contained in Oleoresin: GC-MS instrument (Gas Chromatography-Mass Spectrometry) is a combination of GC and MS tools, this means the sample to be examined first identified by means of GC (Gas Chromatography) new, later identified by means of MS (Mass Spectrometry). GC and MS is the simultaneous combination of force to separate and identify the components of a mixture.

TABLE II
RESULTS ANALYSIS GCMS

Parameters	Extraction Method		
	Ultrasonic	Maceration	Soxhletation
Extraction Temperature	30°C	30°C	60°C
Time Extraction	3 hour	3 hour	7 hour
Components Identified	19	10	8
Oleoresin Components	*Myristicin Eugenol*	*Myristicin*	*Myristicin*
(%)	10,07 2,38	2,32	4,90

Oleoresin form a viscous liquid contained volatile oil cof 15 to 35% volume. Nutmeg oleoresin consist of fatty components, terpenoids, essential oils, resins and aromatic compounds such as myristicin, safrole, eugenol and elemicin [17]. Based on the results of GCMS analysis, the components contained in the oleoresin extraction with ultrasonic waves for 3 hours at 30oC consisted of 19 components with the largest content of aromatic compounds. It were myristicin and eugenol, while the other components consisted of fatty acids (fixed oil). The GCMS identified that the soxhletation number of components was smaller than the maceration and ultrasonic extraction. This because the high temperature extraction.

Based on Table 4.3, it was found that the highest number of components of the oleoresin nutmeg with ultrasonic extraction method 30oC for 3 hours, myristicin obtained 10.07% and 2.38% of eugenol. It is clear that the quality of the extraction of oleoresin produced of ultrasonic extraction was better than the extraction of oleoresin from maceration. The ultrasonic extraction can extracted more components from oleoresin.

IV. CONCLUSIONS

From the results, it can be concluded as follows: Ultrasonic assisted extraction method produced the highest yield at 50oC extraction temperatures with extraction time for 3 hours is equal to 6.2%. Refractive index and density of the oleoresin produced tends to increase over time and temperature of extraction. Soxhlet method requires a longer time and higher temperature to produce oleoresin, while using the maceration and ultrasonic waves assisted extraction methods. It can be performed at a lower temperature conditions in a shorten time. The highest number of identified components of nutmeg oleoresin with ultrasonic extraction method at 30oC for 3hours was myristicin obtained 10.07%. it also obtained 2.38% eugenol. The quality and quantity of a series of extraction method (soxhlet, maceration, and ultrasonic-assisted) has been done, which is consisted in the extraction method with the ultrasonic waves at a temperature extraction of 30^{o} C with extraction time for 3 hours.

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