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SOLVENT EXTRACTION OF GINGER OLEORESIN USING ULTRASOUND

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Abstract

The use of ultrasound in extraction process creates novel and interesting methodologies, which are often complementary to conventional extraction methods. In the present study, the use of ultrasound to extract oleoresin from ginger (*Zingiber officinale* R.) was investigated. The extraction was performed by using ethanol as solvent in the presence of ultrasonic irradiations operating at frequency of 42 kHz at extraction temperature of 60 ^{o}C. The oleoresin extracted was in the form of dark thick liquid with specific ginger flavor. Based on GC-MS analysis, the use of ultrasound was not give an effect on alteration of main component in ginger oleoresin. The main component in extracted ginger oleoresin was zingerone. Gingerol as one of the pungent principle of the ginger oleoresin was not detected due to decomposition of gingerol at a temperature above 45 ^{o}C. Extraction rate of ultrasound-assisted extraction was about 1.75 times more rapid than a conventional system based on soxhlet. The scanning electron microscopy images provided more evidence for the mechanical effects of ultrasound, mainly appearing on cells' walls and shown by the destruction of cells, facilitating the release of their contents.

Keywords: extraction, ginger, oleoresin, solvent, ultrasound

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1. Introduction

Ginger is commonly used as a cooking spice and a medicinal plant throughout the world. It is the rhizome of the perennial plant *Zingiber officinale* in the family *Zingiberaceae*. The ginger plant has a long history of cultivation, known to have originated in Asia and then spread to India, Southeast Asia, West Africa, and the Caribbean. Ginger products, such as essential oil and oleoresin, are internationally commercialized for use in food and pharmaceutical processing. Essential oil is characterized by monoterpenes and sesquiterpenic compounds while the main pungent compounds in the oleoresin are a series of homologues called gingerols and shogaols. In recent years, more and more pharmaceutical effects have been found on ginger. It can act as an aphrodisiac, a carminative, a rubifacient, an anti-asthmatic and as a stimulant to the gastrointestinal tract. Ginger is often used for the treatment of stomachaches, and cardiovascular and motor diseases. It also possesses anti-inflammatory activity and regulates bacterial growth, as well as providing protection for immune-depressed patients, such as individuals who are HIV positive [1].

Many active components have been found in ginger [2]. The active component obtained from ginger is a high value-added product and due to such, there is continued research for improved extraction techniques that will lead to better quality extracts and greater yields. Some classical techniques, such as solvent extraction and soxhlet extraction can be used for this purpose.

Many investigations have reported that a higher extraction yields could be achieved within shorter time period and/or lower temperature when an ultrasound-assisted extraction (UAE) method was adopted for various plant species [3-8]. UAE is a novel method to effectively extract chemical constituents from plant materials. High intensity shock waves generates intense pressures, shear forces and temperature gradient due to the bubble of cavitation inducing the majority of ultrasonic effects within a material, which can produce physical, chemical and mechanical effects [4], making the chemical constituents dissolve in the solvent without heating. Ultrasound can also facilitate the solvatation of plant materials by causing cell swelling and enlargement of the pores of the cell wall. Better swelling will improve the rate of mass transfer, and result in the increased extraction efficiency and/or reduced extraction time [5]. The main advantages of this new method are that the ultrasound can increase the movement of the molecules enhancing the penetrability of the solvent and promoting a higher dissolution rate of plant constituents [6-7] in less time, which may decompose under higher temperatures used in reflux extraction, increasing the extraction yield [3,7-8].

In recent years, there have been several reports on the application of UAE in the isolation of various biologically active compounds from plant materials, for example, release of hemicellulose from buckwheat hulls [3], extraction of sensitive aroma compounds from garlic [9], isolation of essential oil from olives [10], and extraction of phenolic compounds from coconut (*Cocos nucifera*) shell powder [11]. This indicates that the method has potential in the extraction of thermal sensitive constituents used in foods, healthcare products, cosmetics, and pharmaceuticals. Application of ultrasonic extraction of bioactive principles from plants has also been reviewed [12]. In many analytical situations, UAE is an expeditious, inexpensive and efficient alternative to conventional extraction techniques, and, in some cases, even to supercritical fluid and microwave-assisted extraction.

In the present study, we investigated the influence of ultrasound on the extraction of oleoresin from ginger (*Z. officinale* R.), which belong to the Zingiberaceae family. A comparison was also conducted between a conventional extraction method based on soxhlet and UAE.

2. Experiment

Fresh ginger rhizomes were purchased from a local market around Bireun City, Aceh province, Indonesia. Any foreign material such as small stones, sand and plant leaves that may be present after harvesting, transportation and storage was removed. Then the ginger rhizomes were washed, peeled and then cut into uniform sized. The peels were sun dried for more than 2 days when they reached a final moisture content of about 10%, and then they were finely grounded in a laboratory grinder. A stainless steel screen with a mesh size of 2 mm was used to obtain a consistent particle size distribution of ginger powder. The dry plant material was then packed in paper bag and stored at 20 ^{o}C for usage. All other chemical reagents used in the experiments were of analytical grade.

UAE experiments were carried out in ultrasonic cleaning baths Bransonic 8510 with a frequency of 42 kHz and a digital timer to set up time. Schematic diagram of the UAE apparatus used in this study is presented in Figure 1. Powder of 50 g ginger was loaded into a 500 mL flask and 150 mL of ethanol was added as a solvent. The extraction was carried out under extraction temperature of 60 ^{o}C in a specified time. The temperature was maintained using a water bath temperature controller. During experiments with ultrasound, it was also necessary to continuously replace some water in the bath with cold water to maintain temperature. It is also necessary to ensure that the bath water level remained constant during this replacement. Temperature was monitored at the internal extraction vessel and confirmed to be constant within ±1 ^{o}C. After extraction, extract was cooled to room temperature. The extract was clarified by filtration on a water pump, and was then concentrated in a rotary evaporator.

The GC-MS analysis using HP-5MS column with: 30 m × 0.25 mm and 0.25 μm film thickness, were conducted using HP G 1800C Series II GCD system (Hewlett-Packard, Palo Alto, USA). Helium was used as carrier gas. Injector and detector temperatures were 260 °C. The components of the extract were identified by comparison of their mass spectra to those from Wiley 275 and NIST/NBS libraries. The experimental values for retention indices were determined by the use of calibrated Automated Mass Spectral Deconvolution and Identification System Software (AMDIS ver.2.1.),compared to those from available literature, and used as additional tool to approve MS findings. A Hitachi S-3500N field emission scanning electron microscope was used to image ginger particles. A slice of ginger with size of 1 mm thickness was prepared for this purpose. All samples were prepared and analyzed in duplicate.

The conventional extraction was carried out in a similar manner as the one explained for UAE by using soxhlet. This was performed as a control for comparison with UAE.

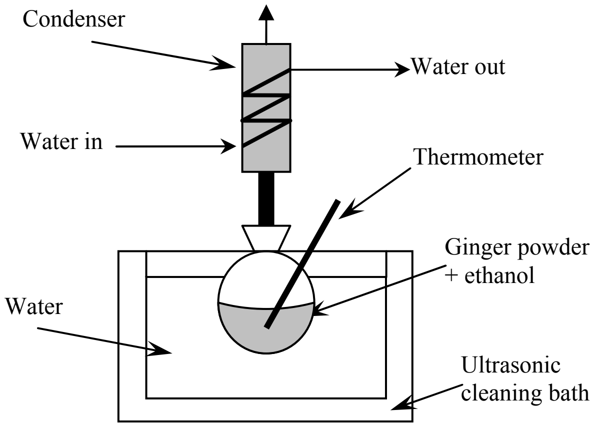


Figure 1. Schematic Representation of UAE Apparatus Used in this Study

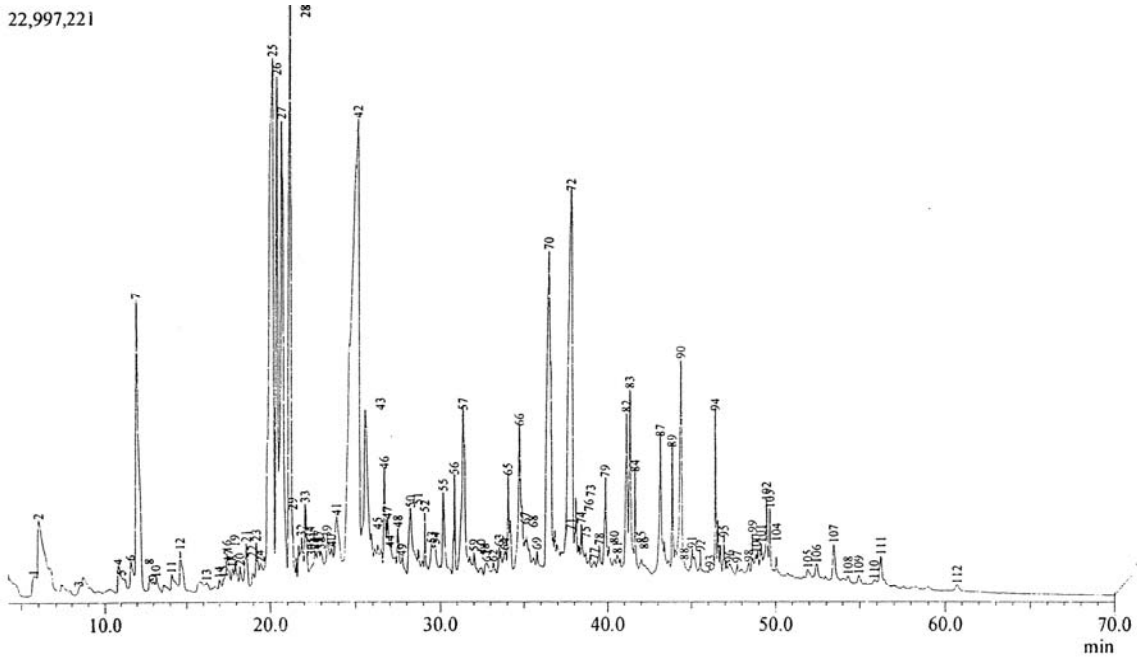


Figure 2. Typical GC-MS Total Ion Chromatogram of Components in Ginger Oleoresin

3. Results and Discussion

In general, the ginger oleoresin obtained from each experiment was very similar in appearance, dark colored, thick liquid and specific ginger flavor. As expected, the composition of the extract was quite complicated with many components resolved using GC-MS. The typical GC-MS total ion chromatogram of components in extracted ginger oleoresin can be seen in Figure 2. Meanwhile, oleoresin components analysis results by GC-MS are presented in Table 1. Only five of the largest component of the composition is presented. Table 1 also shows the results of the soxhlet and the results reported by Zancan *et al.* [13]. The results of GC-MS analysis as shown in Table 1 shows that component zingerone was the largest composition identified either through the extraction by UAE and conventional extraction based on soxhlet. The results of GC-MS analysis also shows that total of 113 and 112 components were identified by soxhlet and UAE, respectively. These results would indicate that the difference in the GC-MS analysis was small, suggesting that ultrasonication did not noticeably influence on changes in components of ginger oleoresin.

Table 1. GC-MS Results of Ginger Oleoresin

Component	Composition (%)	Method
Zingerone	14.47	Soxhlet[a]
Shogaol	7.14	
Farnesene	6.29	
β-sesquiphellandrene	5.44	
Zingiberene	5.13	
Zingerone	13.85	UAE[b]
Ar-curcumene	8.90	
Shogaol	7.92	
Nortrachelogenin	6.74	
β-sesquiphellandrene	5.02	
Zingiberene	16.77	CO_2 Supercritical [13][c]
β-phellandrene	12.86	
Geraniol	9.01	
β-sesquiphellandrene	6.54	
Gingerol + shogaol	3.48	

[a] Extraction time: 420 min, extraction temperature: 78 °C

[b] Extraction time: 300 min, extraction temperature: 60 °C

[c] Extraction time: 420 min, extraction temperature: 35 °C, extraction pressure: 250 bar

However, based on GC-MS result, gingerol as one of the pungent principle of the ginger oleoresin did not detect. This is because gingerol will decompose to zingerone and shogaol at higher temperature (above 45 °C), resulting in the decrease of gingerol contents in the oleoresin [14-15]. In addition, there are several difficulties in the identification and quantification of the gingerols compounds using gas chromatography due to the thermal degradation to shogaols [16]. Meanwhile, another pungent principle of the ginger oleoresin in form of zingerone and shogaol was detected in a significant amount.

Table 2 shows the comparison of oleoresin yield of the extraction process using ultrasonic and conventional extraction of soxhlet. From Table 2, it can be seen that the UAE yield was 7.43% within 240 min, while almost

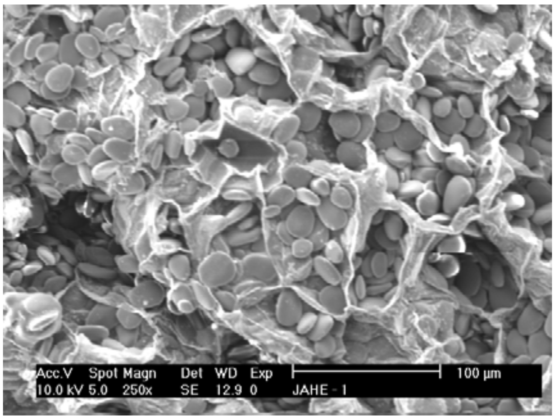
Table 2. Oleoresin Yield of UAE and Soxhlet

Time (min)	Oleoresin yield (%)	
	UAE	Soxhlet
30	5.50	-
60	6.50	-
120	6.80	-
180	7.28	-
240	7.43	-
300	7.81	-
420	-	7.48

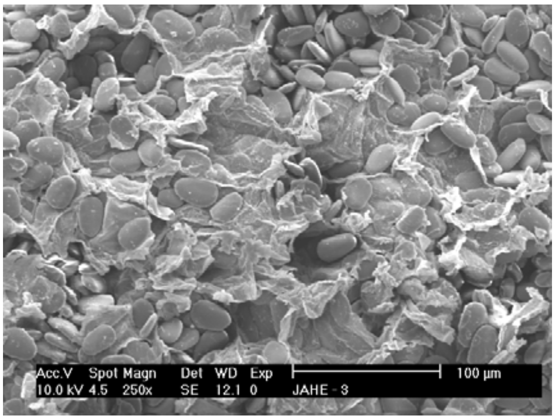
the same yield obtained within 420 min for soxhlet extraction. Extraction rates of oleoresin for UAE were about 1.75 times more rapid than a conventional system based on soxhlet. Also, this indicates that the UAE has time efficiency about 50% compared than soxhlet extraction. Ji *et al.* [17], also states that ultrasound could enhance the diffusion of the solvent in a substance, which the influences of the ultrasound cavity not only around the particles, but also directly to the center of the substance. Meanwhile, Garcia and Castro [18] reported that the ultrasonic extraction time is shorter compared with soxhlet extraction to produce a similar amount of product yield in the process of fat extraction from plant seeds. The same trend was also reported by Ma *et al.* [19] for hesperidin extraction from the *Citrus reticulata* peel.

In general, the longer the time extraction, the higher the yield. This is because a longer extraction time will provide a higher chance of contact between the solvent and material. The solubility of the material will continue to increase with increasing the extraction time until the occurrence of saturation in the solvent. In addition, the longer time of extraction, the more heat is received, and then the diffusion process will increase to accelerate the extraction process. However, as can be seen in Table 2, there is no significant change of oleoresin yield obtained for the extraction time above 60 min. This might be due to the influence of ultrasonic was maximal at the time of extraction approximately 60 min, so the change in time extraction did not give a significant change of yield. According to Balachandran *et al.* [20], the use of ultrasonic will increase the effective diffusivity in the mass transfer process and this effect will be optimum in a short time of process.

To gain further insight into the effect of ultrasonic vibration on the physical structure of ginger particles, scanning electron microscopic (SEM) images of the plant cells after extraction was obtained to provide visual evidence of the disruption effect. Figure 3 shows a set of SEM images of ginger particles at a magnification factor of 250×. As shown in Fig. 3(a), experiment result shows that ultrasonic vibration will caused the disruption of the cell structures and increase in the accessibility of the solvent to the internal particle structure, thereby facilitates removal of the cell contents. It will also enhance the intra-particle diffusivity [14]. Meanwhile, as presented in Fig. 3(b), the SEM images of experiment without ultrasound also show similar phenomena; however, the disruption of the cells occurred in a smaller scale.



(a)



(b)

Figure 3. Scanning Electron Microscopy Images of Ginger Particle (a) Experiment with Ultrasound; and (b) Experiment without Ultrasound

Nevertheless, additional studies will be required to quantify the contribution of the individual effects of high-intensity ultrasound and extraction parameters such as temperature, pressure and solvent type on oleoresin yield and to gain a better understanding of the mechanism of UAE.

4. Conclusion

Ultrasound has the potential to be used in oleoresin extraction processes to reduce processing time. Results were attributed to mechanical effects due to ultrasonically induced cavitation increasing permeability of plant tissues. Gas chromatography analysis of ultrasonicated ginger oleoresin did not show significant changes in main components of ginger oleoresin. However, gingerol as one of the pungent principle of the ginger oleoresin did not detect because gingerol will decompose to zingerone and shogaol at a temperature above 45 °C. Scanning electronic microscopy analysis was carried out on the ginger slice after the extraction. The images are powerful evidences to show the effect of ultrasound. Our study suggests that high-intensity ultrasound may reduce time required to extract oleoresin from plant sources and hence improve throughput in commercial oleoresin production processes.

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