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The Effect of Ultrasonic Pre-treatment on Yield of Hydrodistillation Cardamom (*Amomum Cardamomum*) Seeds Essential Oil

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Abstract. *Amomum Cardamomum* can be processed to obtain the essential oil, which has numerous benefits, including an active antimicrobial and antioxidant. Hydrodistillation is used to extract oil from cardamom seeds with the help of ultrasonics as a pre-treatment that provides higher effectiveness and efficiency. The purpose of this research is to investigate the effect of pre-treatment sonication on the hydrodistillation method. The research was carried out by performing conventional hydrodistillation and ultrasonic assisted extraction followed by hydrodistillation (UAE-HD) with amplitude (30, 60, and 90%) of the pre-treatment sonication was varied as the main parameter and as a factor affecting the extraction process in two repetitions. The results showed that UAE-HD significantly increased the yield of essential oil while also shortening the extraction time. With a predicted optimum process time of 158.25 min, the 90% amplitude obtained the highest yield of 6.97% (v/w). Results from the GCMS analysis showed cardamom essential oil component compounds such as α-pinene, β-pinene, p-cymene, D-limonene, γ-terpinene, α-terpeniol, terpinyl acetate and 1.8 cineol with the highest percentage (>60%) as the major components.

Keywords: Ultrasonic, hydrodistillation, cardamom oil, essential oil, cineol.

INTRODUCTION

Amomum cardamomum is a spice producer with high economic values. Cardamom is widely used in the food to pharmaceutical industries. Cardamom is included in the nine major world spices commodities and is traded in the form of essential oils and dried fruit. In the health sector, cardamom has antihypertensive benefits with a diuretic effect, prevents chronic disease, overcomes digestive problems, is antibacterial, and its cytotoxic properties can fight cancer cells. Cardamom contains essential oils, fats, pigments, proteins, cellulose, sugars, starches, and minerals, with the largest component being starch. The highest fiber content is contained in the skin, up to 31% [1]. According to Hartady (2020), cardamom seeds contain 60 - 80% cineole consisting of several types, and the rest contain α-pinene, β-pinene, camphene, limonene, ρ-cymene, α-terpineol and α-humulene and 2,2'-methylene bis [6- (1,1-dimethyletyl) -4-ethyl] phenol. Essential oil from cardamom seeds is used in various industries such as perfume, food, and

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pharmaceuticals. In addition, the community uses the seeds and rhizomes as traditional medicine in treating various diseases [2].

Cardamom seed essential oil can be extracted through a variety of methods. The most popular and cost-effective extraction method for obtaining essential oil from plant sources is hydrodistillation. This process technology is a conventional method that requires a lot of time, but ultrasonic assisted extraction (UAE) only takes a few minutes to hours. Ultrasonic pre-treatment has been known by the term sonication to improve the efficiency and quality of the product. Sonication has been shown to increase yield as well as reduce energy consumption. According to Cao (2018), in the case without ultrasonic treatment, the total energy consumption was 6.275×106 J/g and with ultrasonic treatment the minimum energy consumption was 5.175×106 J/g. These results show that ultrasonic treatment can reduce 5-19% energy consumption when freeze drying barley grass samples [3].

Ultrasonic Assisted Extraction (UAE) technology is a new extraction technology developed as an alternative method, where ultrasonic waves are used to help the extraction process. Panda & Manickam (2019) review shows that ultrasonic extraction can extract essential oils in 15-30 min compared to 6 h with conventional extraction methods. Furthermore, UAE can reduce the degradation of essential oil components caused by high temperatures, resulting in higher yields. Azwanida (2015) also reported that cell disruption and effective mass transfer are two main factors that improve the results of using ultrasonic waves [4,5].

The use of ultrasonics on plant materials produces both physical and chemical effects. Some of the physical effects of ultrasonics include sono-capillary effect (increased solvent penetration into cell membrane), sonoporation (cell content release to solvent), and detexturation (damage to plant cells structure) [6]. Some research states that ultrasonic extraction increases the gallic acid yield of Labisia pumila by 1.23 times compared to conventional methods at the same time [7] and research by Ramli, Ismail, & Rahmat (2014) provided that ultrasonic assisted extraction provides the highest yield of dragon fruit (90.08%) compared to conventional methods (73.27%). The conventional method takes two hours to extract the entire component, while UAE takes only 30 min [8]. UAE technology application is expected to improve the yield and quality of cardamom essential oil. The objective of this research was to evaluate the effect of amplitude ultrasonic as a pretreatment cardamom seeds on the hydrodistillation method (UAE-HD) compared to conventional hydrodistillation (CHD).

MATERIALS AND METHODS

Javanese cardamom (*Amomum cardamomum*) is obtained from Padang city with Tusanco brand. Cardamom seeds was separated from the skin, then chopped by grinder and sieve until 8 mesh of particle size. The manner of hydrodistillation process was placed of cardamom seeds (100 g) in a 3L round flask and added 2L of aquadest. Hydrodistillation using Clevenger apparatus is carried out for 6 hours until cardamom oil was taken. The product of essential oil in distillate with the oil layer is separated from water. Remove impurities of water using sodium sulfate, then placed in a vial bottle in keep the oil in refrigerator at 10°C. Hydrodistillation were performed in duplicates experiments and the average value of the extraction result was recorded.

The experiment using Ultrasonic Assisted Extraction (UAE) by sonicator with high power amplitude (Qsonica Sonicator 1375) and probe size of 1" (25 mm) was conducted following Morsy's method (2015) with modification [9]. 100 g of Cardamom seeds powdered was weighed and placed in beaker glass 3L. Sonication extraction using solid feed ratio (SFR) was measured of 1:20 (b/v). The ultrasonic probe was dipped in a mixture with a depth of about ± 3 cm. The pretreatment step was isolated by aluminum foil and placed in an ice bath. Ultrasonic Assisted Extraction uses 10% of the maximum power output (137.5 W) through energy conversion calculations. The variable process of ultrasonic were studied to obtain the high yield of cardamom oil follow on **Table 1** in below. The product of oil was analyzed component using GCMS instrument with Agilent 15977 system with equipped of Agilent 7683B Series auto-injector, coupled to an Agilent 5975 Inert Mass Selective Detector. The carrier of gas helium used 40 ml/min and DB 5 of column.

TABLE 1. Variable process of ultrasonic as pretreatment process

Amplitude and Energy	30% (120 KJ); 60 % (195 KJ); 90% (250KJ)
Time	15 min
Temperature	20°C
Pulse	0

RESULT AND DISCUSSION

The effect of extraction method on the total extraction time, yield and chemical composition are summarized in **Table 2** and illustrated in **Fig. 2**. Sonication pre-treatment accelerates extraction time and increases the yield of essential oils. Conventional hydrodistillation takes 5 h to extract the entire essential oil from cardamom seeds, while the combination of hydrodistillation and sonication pre-treatment cuts the extraction time (≤ 3 h). UAE-HD significantly delivers a higher yield of essential oils compared to conventional HD (CHD). The results was similared with research by Morsy (2015), which concluded the combination of UAE-HD (ultrasonic assisted extraction followed by hydrodistillation) method lasted very quickly (< 1 h) while 6 h was needed to extract all essential oils from cardamom seeds and yield of essential oil increased significantly [9].

TABLE 2. The effect of extraction method

Parameter (%)	Rf	Method			
		CHD	**UAE-HD 30%**	**UAE-HD 60%**	**UAE-HD 90%**
α-pinene	8.486	2.79	1.54	2.37	1.77
β-pinene	9.872	9.58	6.09	6.27	7.66
p-cymene	11.372	1.23	4.49	1.48	0.92
D-limonene	11.498	5.51	10.15	4.65	5.69
1,8-cineole	11.687	66.86	67.53	68.68	69.43
γ-terpinene	12.405	2.07	1.25	1.12	1.87
α-terpeniol	16.564	2.54	1.99	2.59	2.58
Terpinyl acetate	20.773	2.99	2.28	3.11	2.79
Other components		6.43	4.68	9.73	7.29
Total volume (mL)		5.25	6.5	6.75	7.2
Total yield (%) (w/w)		4.01	5.45	6.35	6.97
Optimum extraction time (min)		322.14	183.25	172.75	158.25
Total ultrasonic energy (KJ)			120	195	250

Pre-treatment sonication with an amplitude of 90% gives the largest yield of essential oils until 7% compared to others, which indicates that the magnitude of the amplitude has an impact with the increase in yield of essential oils. Waves generated by transducers converting electric power to mechanical energy will propagate through propagation media to the seed of cardamom via cavitation. Acoustic streaming and cavitation are caused by the intrusion of ultrasonic waves into the extraction process.

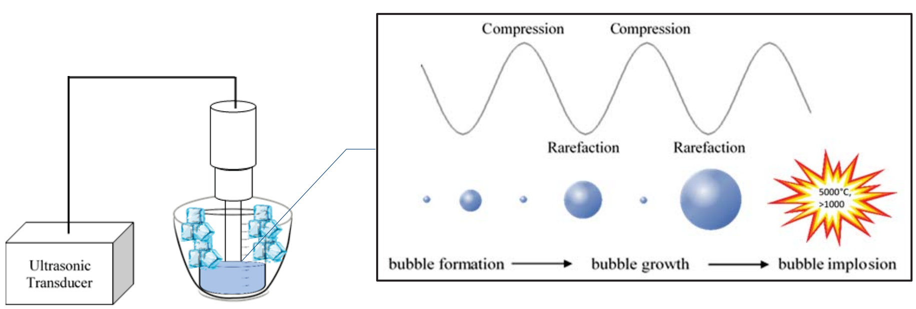


FIGURE 1. Cavitation Mechanism in Ultrasonic Pre-treatment [9].

The presence of acoustic streaming helps in the maximization of the cavitation phenomenon. The collapsed of cavitation bubbles affect the rate at which particle collisions become high, causing cell wall destruction. As a result, it is easier to free the components of compounds contained in seed cells. Cavitation creates a fast-moving solvent flow through the surface cavity of the seeds, causing damage to the outer surface peeling as well as particle damage. During the sonication process, cavitation phenomena will occur repeatedly in a short period of time, maximizing solvent penetration against seed.

During the sonication process, cavitation phenomena will occur repeatedly in a short period of time, maximizing solvent penetration against seed. According to (Hashemi et al., 2016), the higher the ultrasonic amplitude passing through a medium, the more cavitation bubbles will disperse on the surface of the solid particles membrane, resulting in larger micro factories [10]. The release of essential oils from cardamom seed cells is assisted by destruction to the cell walls of cardamom seeds caused by cavitation phenomena. When ultrasonic probes generate longitudinal waves as ultrasonic waves interact with solvents, they produce alternating compression and rarefaction areas between the molecules. In this area, cavitation bubbles form, causing pressure changes. The bubble attaches to the sample's surface, and when it collapses, they break down the cell wall and the chemical components inside it are transferred into the solvent. As a result, the yield of essential oils from cardamom seeds has increased. Some of the advantages of using pre-treatment sonication involve increased mass transfer, cell breakdown, increased solvent penetration, capillary effect, high yield, and short extraction time [11].

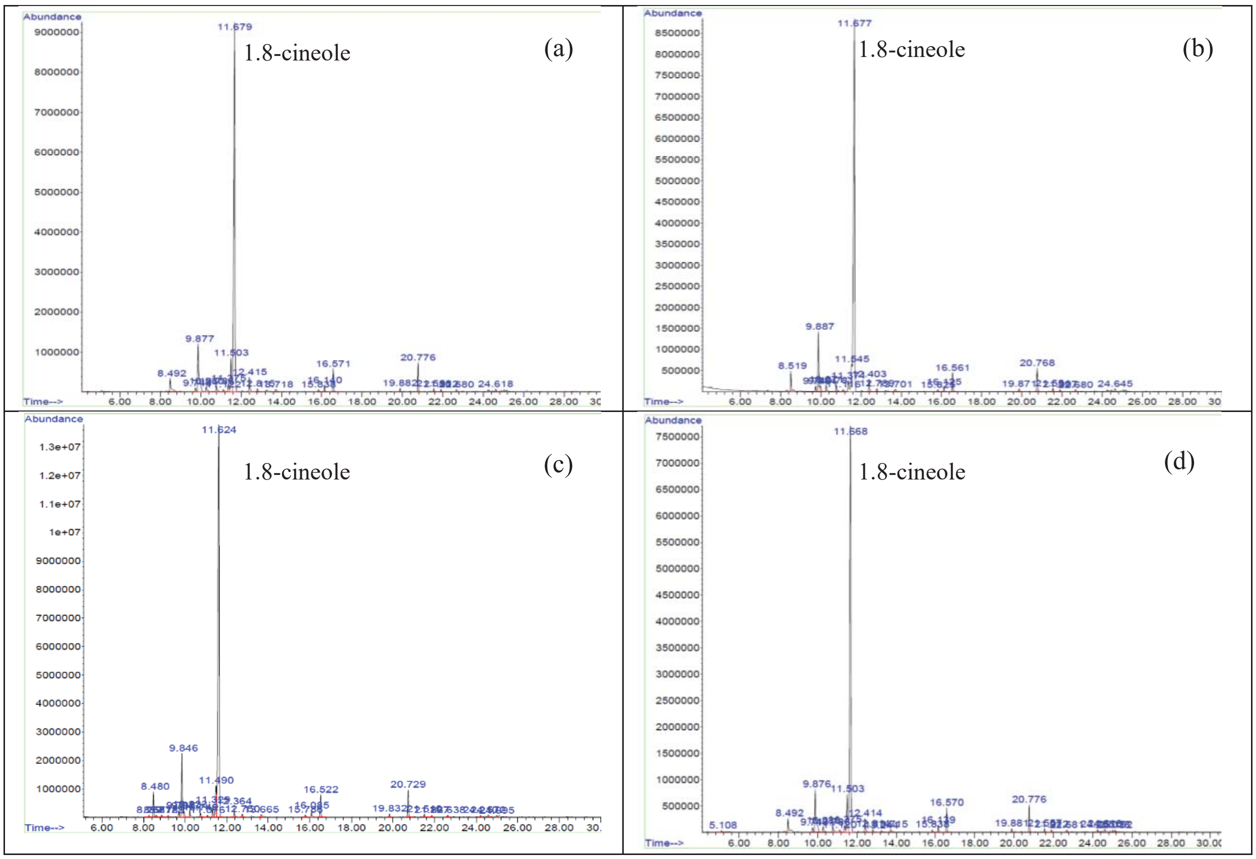
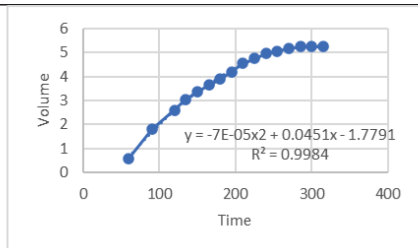
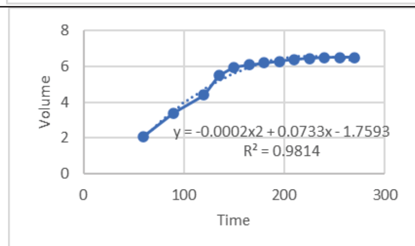
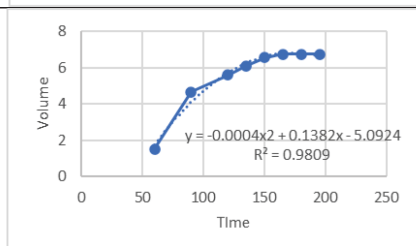
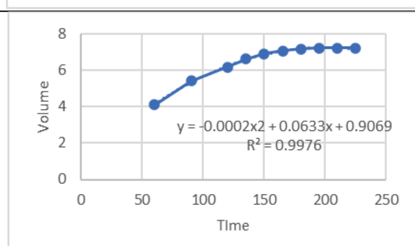


FIGURE 2. GC-MS chromatogram of cardamom essential oil by (a) CHD (b) UAE-HD 30% (c) UAE-HD 60% (d) UAE-HD 90%

Figure 2 showed that the identify of 26 cardamom essential oil compounds, including α-pinene, β-pinene, p-cymene, d-limonene, 1,8-cineole, γ-terpinene, α-terpeniol, dan terpinyl acetate. Ratri et al. (2020) identified six components of java cardamom essential oil compounds: 1,8-cineol, camphene, terpinen-4-ol, a-terpinol, terpinyl acetate, and eugenol [12]. The major component of cardamom essential oil is 1.8-cineole, which contributes for more than 60% of the total content. The use of ultrasonic pre-treatment increased the percentage of 1.8-cineole to 69.43% with a 90% amplitude. This is a significant improvement over Rosmainan (2017), which only contains 64.15% of 1.8-cineole. The increase in 1.8-cineole levels is proportional to the increase in cardamom essential oil yield, which is caused by the breakdown of the cardamom seed cell layer, allowing the 1.8-cineole compound to be fully extracted [13].

The trendline curve is an approach to describing the relationship between time and volume using a second-order polynomial equation. The R^2 valued at more than 0.98 indicates that this approach can be validated with high precision for approaching data. A calculus solution based on a second-order polynomial equation is required to determine the optimum time (x-axis) where the maximum yield is achieved (y-axis). The derivation of the second-order polynomial regression is used to solve the equation. In order to be solved, the equation's derivation must be equated with zero [14]. The optimum time illustrated in **Table 3** is the value X, which is derived from the derivation of the second-order equation. There are very clear for the optimum time process of hydrodistillation with pretreatment by ultrasonic various amplitude compared to conventional method. The results of time process were obtained of 332.14 min for conventional HD (CHD) and various amplitude UAE-HD 30%, UAE-HD 60% and UAE-HD 90% were showed that 183.25; 172.75; and 158.25 min, respectively. For the Table 3, the best time process of hydrodistillation was performed in UAE-HD 90%.

TABLE 3. Calculation of the optimum time

Method	Trend Line	Calculation
CHD		$y = -7E\text{-}05x^2 + 0.0451x - 1.7791$ $y' = 0.0451 - 0.00014x$ $0.00014x = 0.0451$ $x = 332.14$
UAE-HD 30%		$y = -0.0002x^2 + 0.0733x - 1.7593$ $y' = 0.0733 - 0.0004x$ $0.0004x = 0.0733$ $x = 183.25$
UAE-HD 60%		$y = -0.0004x^2 + 0.1382x - 5.0924$ $y' = 0.1382 - 0.0008x$ $0.0008x = 0.1382$ $x = 172.75$
UAE-HD 90%		$y = -0.0002x^2 + 0.0633x + 0.9069$ $y' = 0.0633 - 0.0004x$ $0.0004x = 0.0633$ $x = 158.25$

Ultrasonic wave propagation is affected by sonication parameters: power input, sonication time, probe diameter, and sonication frequency [15]. One type of ultrasonic application that is frequently used in the field of chemistry is the ultrasonic probe type. As it has used a modified probe, the ultrasonic horn allows for power control. The energy produced by amplitudes of 30, 60, and 90% is 120 KJ, 195 KJ, and 250 KJ, respectively. Energy (Joule) is directly proportional to the square of the amplitude so that with the greater the amplitude used, the energy used or produced by the sonicator will be greater as well [16]. According to Morsy (2015) reference, the energy generated fulfills 10%

of the maximum power output of 137.5 W from Qsonica Q1375 (1375W power output) through energy conversion calculations. The use of 90% amplitude produces maximum energy that is still within 10% of the maximum power output (250 KJ), ensuring that the energy produced does not exceed the limit or consume excessive energy [9]. **Figure 3** was shown that the relationship of time process hydrodistillation and volume of cardamom oil with various amplitude of ultrasonic compared to conventional technique.

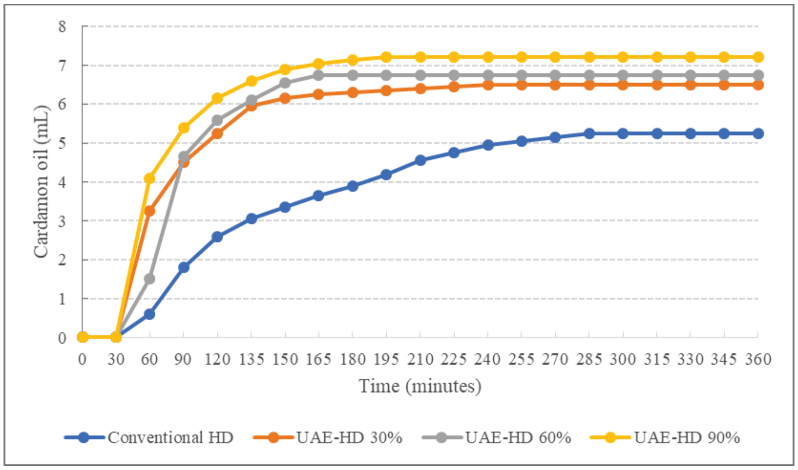


FIGURE 3. The relationship of time process hydrodistillation and volume of cardamom oil with various amplitude of ultrasonic compared to conventional technique.

According to the graph, ultrasonic asssited extraction results in the release of the same essential oil as conventional hydrodistillation yields in just 120 min. Meregalli et al. (2020) research was found that using ultrasound, Anthocyanins achieved the same result (120mg/100g) in 90 min, while conventional methods took 120 min [17]. The higher the sonication amplitude, the shorter the extraction time will be. This can be explained that sonication probes generate exothermic heat that is proportional to the higher amplitude. Ultrasonic extraction with exothermic heat causes the diffusion process to increase and the extraction process to be accelerated, resulting in a shorter extraction time [4].

CONCLUSION

Combining ultrasonic as pretreatment and hydrodistillation to obtain the cardamom oil was shown successful in reducing the time process of hydrodislllation. Compared to conventional hydrodistillation, the UAE-HD gave the time process of hydrodistillation increase 1.5-2 times. The best condition UAE-HD were obtained in 90% of amplitude, and highest yield of essential oils of 7%. The componen analysis showed that the cardamom essential oil gave α-pinene, β-pinene, p-cymene, D-limonene, γ-terpinene, α-terpeniol, terpinyl acetate and 1.8 cineol with the highest percentage (> 60%) as the major components. The breakdown of large cavitation bubbles in the seed cell membrane will occur as the amplitude of sonication waves increases, facilitating the release of oil from the cell and increasing mass transfer. Compared to conventional hydrodistillation, the impact has an effect on the yield of cardamom seed essential oil and the short extraction time.

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