Enhancement of Javanese Cardamom Fruit Seed and Peel Oil Yield by Steam Distillation

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Abstract

The main objectives of this research are to extract essential oil from various parts of cardamom by steam distillation, study the effect of physical properties of cardamom (size and moisture content), study the effect of extraction conditions (distillation time, steam pressure, condensate flowrate) on oil yield, the resulting essential oil was determined by Gas Chromatography (GC). To obtain the maximum yield, distillation time, pressure, and condensate flow rate were identified as potential critical process parameters by reviewing the literature. Those three parameters were then investigated by single-factor experiments. The essential oil yield first increased and decreased with the extension in the distillation time. It is thought that increasing pressure at a constant temperature increases the distillation rate due to the high possibility of separation of organic matter, but the essential oil yield would be then decreased with the increased pressure (> 1 bar). Condensate flow is closely related to the steam flow rate which in turn affects the heat that is transferred toward the plant material and the steam pressure inside the distillation tank when it is being used. This exciting result opens the possibility of developing an economic essential oil with high efficiency from the abundant Indonesian herb resource.

Keywords: *Amomum compactum, essential oil, extraction, 1,8-cineol*

Introduction

Indonesia has two types of Amomum compactum, including false cardamom or Java cardamom (*A. compactum Soland ex Maton*) and true cardamom (*Elettaria cardamomum (L.) Maton* or *A. cardamomum L.*). Java cardamom (A. compactum Soland ex Maton) has been used more often than other types. Many people in the world use herbs, especially cardamom for various purposes. Not surprisingly, cardamom, the "Queen of Seasoning", is not only known as one of the most expensive herbs, but it also has various benefits such as being easily found in nature, having special tastes and odors, and containing high bioactive compounds.

Previous research has reported on the phytochemical compounds of Java cardamom. The essential oil contents of Java cardamom range from 3.30–4.52% for seeds and 0.99–1.08% for leaves [1]. As far as the authors know, the essential oil contents of Java cardamom fruit, which is a combination of peel and seed, have not been observed yet. Therefore, in this study, we evaluated and compared the essential oil yield from various part of Java cardamom fruit by steam distillation. Essential oils are volatile natural and complex mixtures containing many unique compounds, affecting unique benefits [2]. Essential oils from different plant organs within the same plant have been studied [3-5].

The principle behind steam distillation is that it enables a compound or mixture of compounds to be distilled at a dependent-temperature, lowering the boiling points of the compounds. Essential oil consists of volatile compounds with boiling points higher than 200 °C. In the presence of steam or boiling water, however, these substances are evaporated at 100 °C at atmospheric pressure [5]. The implication is that the extraction rate of essential oils could be influenced by temperature while excessive heat may degrade the quality of the extracted essential oil [6].

Another parameter that could affect the yield is the physical properties of raw material, such as moisture content and particle size [7-9]. Caputo [10] reported that the drying process can affect the essential oil content. In general, small particle sizes of the plant raw materials favours increase in yield of the extraction, however, this variable depends on time distillation. Therefore, we evaluated the effect of moisture content and size of cardamom as a raw material on essential oil yield. Moreover, the objective of the present research work was to evaluate the effect of pressure and distillation time on the yield of the essential oil from cardamom. These parameters were aimed to increase the yield of essential oil. The chemical component of the obtained essential oil was evaluated by GC-MS.

Experimental section

Materials

Java Cardamom (*Amomum compactum* Soland ex Maton) was supplied from Sukabumi, Jawa Barat, Indonesia. The moisture content of Cardamom was reduced to less than 12% by drying at room temperature. N-hexane, toluene, phenolphthalein, 0.1 N KOH, 0.5 N HCl, and 90% ethanol were purchased from Sigma-Aldrich. Distilled water was self-made in the laboratory.

Preparation and characterization of cardamom

First, the cardamom fruits were washed with distilled water to remove foreign matter. After washing, the clean cardamom fruits were dried to a moisture content of less than 12%. The dried cardamom fruits were then grinded and sieved to a different size 1 mesh (>19 mm), 2 mesh (9.52), 4 mesh (4.76 mm), 8 mesh (2.38 mm), 14 mesh (1.41 mm). Proximate parameters (moisture content, ash content, fat content, protein, and carbohydrates) were evaluated using standard techniques.

Distillation process and characterization of essential oil

The fresh cardamoms were dried at room temperature until their weight was constant and their water content was calculated. For each sample, 5 kg of dried cardamoms with different parts were extracted by steam distillation for 4, 6, and 8 hours. Steam distillation was employed to extract the essential oil from cardamom fruit (a mixture of seed and peel). The obtained oils were kept in labeled bottles and were stored in a fridge before analysis. The oil yields were expressed in % (w/w) of the dry weight of cardamom.

Quantification of volatile compounds in cardamom essential oils was performed by Gas Chromatography-Mass Spectrometry (GC-MS Shimadzu, QP 2010). Helium was used as the carrier gas at an initial flow of 1mL min^{-1}. The oven column temperature was set at 70 °C and the temperature for injection was set to 310 °C. GC-MS analysis was performed with a retention time of 50 minutes. MS was employed in EI mode at 70 eV coupled with Shimadzu 2010 GC. Chemical compounds of extracted cardamom oils were quantified by the relative peak area from a chromatogram and chemical components were identified by comparing the mass spectral pattern and retention time with the NIST 147 database library.

Results and discussion

Proximate analysis of cardamom Fruit

Nutrients, such as protein and carbohydrates, should be assessed to contribute to the optimization of medicinal plants [11]. In recent years, a lot of research has been performed on producing and evaluating various sources of proteins and fibers [12]. Proteins consisting of essential amino acids have nutritional values for human health. Besides using such plants as medicine, other nutrients such as fat, protein, fiber, nitrogen, and carbohydrates could be essential for dietary intake [13]. Table 1 describes the proximate analysis results of the cardamom. Cardamom had a high carbohydrate content (68%), followed by ash content (16%) and protein (7%).

Table 1 Proximate compositions of cardamom fruit

Composition	Percentage dry weight basis
Moisture content	6.9 ± 0.18
Ash content	16.4 ± 0.28
Fat content	0.9 ± 0.09
Protein	7.5 ± 0.34
Carbohydrate	68.4 ± 0.15

Similarly, Amma et al reported a protein value of 12.7%, moisture 9%, and ash 6.97% in cardamom [14]. This difference in functional properties may be related to environmental conditions, particularly in the role of ameliorated soil and climatic conditions of the area [15-17]. The proximate composition of cardamom plays a decisive role in assessing its nutritional significance. The large amounts of ash that were obtained, make cardamom a promising sources of minerals.

Effect of various parts of cardamom

The essential oils from different parts (seed, peel, fruits) of cardamom were obtained by Steam Distillation. The yield percentage of the different essential oils obtained with steam distillation methods are presented in Fig. 1. Based on this data, it can be seen that the part of cardamom producing the highest yield of essential oil is the seed (4.18%), followed by all part of cardamom (2.26%) and peel (0.89%). Similarly, Agoes [18] reported that the most important part of cardamom is the seeds, which produce the highest essential oil (3-7%). Moreover, a different part of cardamom fruit has been investigated, resulting in the low yield of essential oil when it was produced from cardamom pod and peel which was 0.2% and 0.18%, respectively [19]. The reason may be that the component of fruit, seed, and peel of cardamom was different. Cardamom fruit consists of peel and seed.

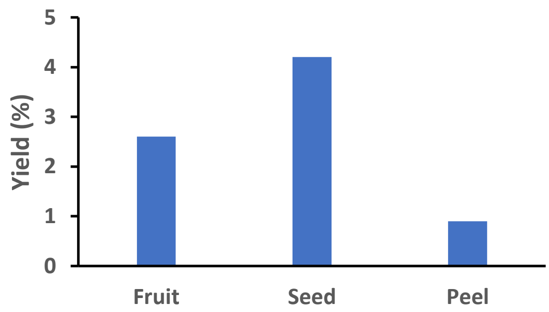

Fig. 1 Yields of essential oils from different parts of cardamom

Although cardamom seed is the most valuable part of producing essential oil, it will take time to extract essential oil and increase the production cost to extract the seed from the fruit. Moreover, the seed of cardamom is only 20% of fruit, leading to the utilization of seed fruit as a source for the essential oil not effective. Therefore, in this study, fruit would be used as a source for the production of essential oil.

Effect of size of cardamom fruit

In general, the smaller size of raw material produced more essential oil yield compared to larger ones. However, there are other factors affecting the essential oil yield, such as its volatility and distillation time in the absence of an airtight collection system. For very volatile essential oils, the longer the distillation time, the lesser the amount of essential oil to be yielded. Using a larger size of raw material is one way to invert this trend.

Another observation in agreement with the literature is that a particle size of 9.52 mm yields the largest amount of essential oil whereas the cardamom size of 1.41 mm produced the smallest yield as clearly shown in Fig. 2. Clearly, the values of 2.28, 3.47, 3.37, 2.57, 0.82 essential oil yields are recorded at particle sizes of 1 mesh (>19 mm), 2 mesh (9.52), 4 mesh (4.76 mm), 8 mesh (2.38), 14 mesh (1.41) respectively. This trend could be explained in terms of the surface area of cardamom: the large particle sizes have more surface areas filled with essential oil than cardamom with a small surface area. Therefore the small particle sizes will yield more oil at shorter distillation time but as the time increases, the oil yield will decrease as most of oil cells are used. In contrast, the large particle sizes will produce lesser oil at the beginning of the distillation but have large surfaces to continue yielding more oil with an increase in distillation time. This agreed with the findings of Kiriamiti et al [20] which explained that extraction yield could be affected by the changes in the mean particle size.

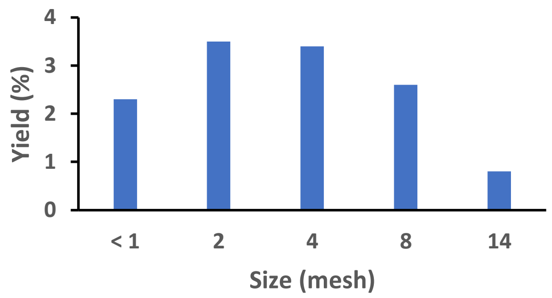

Fig. 2 Yields of essential oils from different sizes of cardamom fruit

Effect of moisture content of raw material on essential oil yield

In general, essential oil yield increased after drying. However, based on this result, the lower the moisture content of raw material, the lower the yield of essential oil (Fig. 3). The moisture content of 11% showed the highest essential oil (3.47%), whereas the driest raw material showed the lowest yield of essential oil (2.91%). It was also depicted that essential oil increases proportionally up to the moisture content of 11% only; further drying of the sample towards less moisture content (9%) did not increase the essential oil. It is inferred from the data that drying up to 11% moisture content is sufficient for the extraction of essential oil.

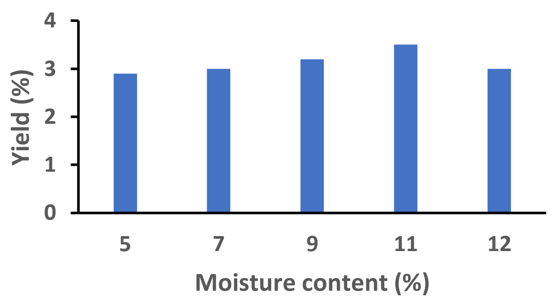

Fig. 3 Yields of essential oils from various moisture content of cardamom fruit

Determination of optimum distillation conditions

The optimal distillation conditions were decided upon by using various distillation times, pressures, and steam flow rates. The experimental variables were optimized to obtain the maximum yield of essential oil at a defined temperature. The temperature for distillation was set to 125 °C, to make sure all the essential oil had been successfully extracted.

The essential oil from cardamom fruit and seed in this study yield of 3.08%. The yield of cardamom oil obtained from distillation time of 4, 5, 6, 7, and 8 hours was different, however, the yield of essential oil tends to increase with the increasing of distillation time. On the other hand, distillation times 6, 7, and 8 hours gave no significantly different yield of essential oils. The highest yield of essential oils was obtained from distillation time of 7 hours and the essential oil yield decreased with increasing the length of distillation time. This is probably due to the volatile oil yield, which first increased and then decreased with the extension in the distillation time.

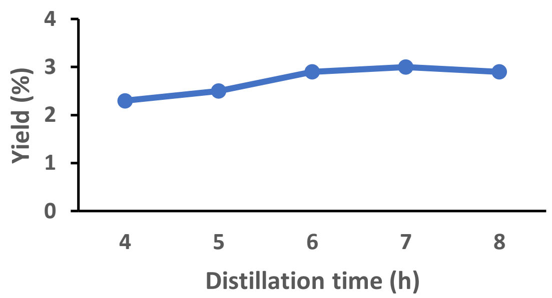

Fig. 4 The effect of distillation time on cardamom essential oil yield

The effects of various pressure and flow rates on the essential oil extract were studied under conditions at 0.5–1.5 bar and flow rate at 12-42 mL min^{-1}. It was observed that there were no significant differences in the amounts of extracted oils. At pressures of 1 bar, the yields of oil extract (3.3%) were higher compared to 0.5 and 1.5 bar. In general, it is thought that increasing pressure at a constant temperature increases the distillation rate due to the high possibility of separation of organic matter. However, in contrast, Fig. 5 showed that the essential oil yield would be decreased with the large pressure (> 1 bar). Passing steam through organic matter in the extraction process is an important thing to condense and form a mixture of steam and matter. However, the excess steam passing through this mixture leads to the evaporation of the required organic compounds as a part of the mixture. From this result, we concluded that 1 bar is the effective pressure to obtain maximum oil yield.

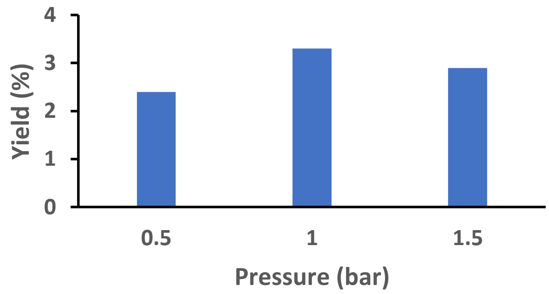

Fig. 5 The effect of pressure on cardamom essential oil yield

The amount of essential oils can be influenced by the condensate flow rate in the steam distillation process. Fig. 6 revealed the amount of essential oil obtained at various condensate rates. It was shown that increasing the condensate flow rate from 12 mL min^{-1} to 18 mL min^{-1} improved slightly the essential oil yield. The highest essential oil content (3.47 ± 0.30%, v/w) was obtained when the condensate rate was set at 18 mL min^{-1} for 420 min. Condensate flow rate is closely related to the steam flow rate which in turn affect the heat that could be transferred toward the plant material and the steam pressure inside the distillation tank when it was being used. It led to more energy transferred into the plant material due to higher temperature input. There is a synergistic effect to improve the system's ability to distill essential oil from the surfaces of plants, also releasing the oil from untouched oil reservoirs and forcing it to diffuse through cardamom particles. It makes the initial oil distillation rate increase which in turn increases the final essential oil yield. In addition, non-heat-resistant components can also be easily degradable during prolonged heating, reducing the yield of essential oil at a lower steam flow rate [21-23]. However, on the contrary, Fig. 6 shows that the yield of oil extract was obviously increasing but the essential oil yield would be then decreased in the high condensate flow rate.

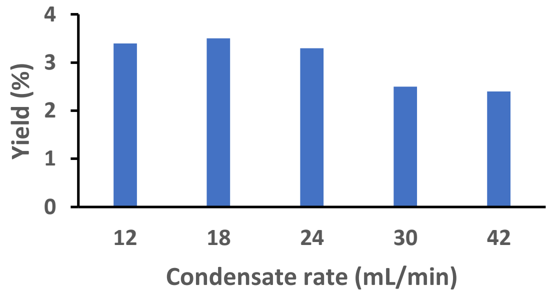

Fig. 6 The effect of pressure on cardamom essential oil yield

The yield obtained at high distillation rates (3.3%, 2.5%, and 2.37% at three operating conditions of 24, 30, and 42 mL min^{-1}, respectively) showed lower values compared to the essential oil content at 18 mL min^{-1}. On the other hand, the rate at the lower limit of 12 mL min^{-1}, had a lower efficiency (3.42 ± 0.23%) compared to those at the higher rate of 24–42 mL min^{-1}. It could be explained by the inadequate dissipation of energy needed to extract the essential oil and the degradation of thermally unstable components when exposed to large amounts of heat. Therefore, a condensate rate of 18 mL min^{-1} for distillation was chosen for further studies.

Essential Oil Component

The obtained essential oil component was analyzed by gas chromatography, as shown in Fig. 7 and Table 2. At least four compounds were identified from the essential oils of cardamom fruit by steam distillation under the condition of 152 °C and 1 bar. Based on this result, the main chemical component of the essential oil was *1,8-cineole* (69,39%), while the contents of β-pinene (9.24%), α-*pinene* (3,31%) and α-*terpineol* (3,56%) were the minority. According to Alkandahri and co-workers, *A. compactum* consists of c.a. 60-80% cineole and other components such as α-pinene, β-pinene, camphene, limonene, ρ-cymene, α-terpineol and α-humulene [24]. It indicated that the main product component has the characteristic of a low boiling point, water-insoluble, and low molecular. Similarly, Amma et al [14] reported that the yield of essential oil by steam distillation is low, extracting low molecular, water-insoluble substances with low boiling point. This result was confirmed by a decrease in essential oil yield with the extension in the distillation time (Fig. 4) and the increase in steam pressure (Fig. 5). A longer duration of extraction slightly decreased essential oil in the case of after 7h, but there was significant difference between yield essential oil with steam distillation across all steam pressure.

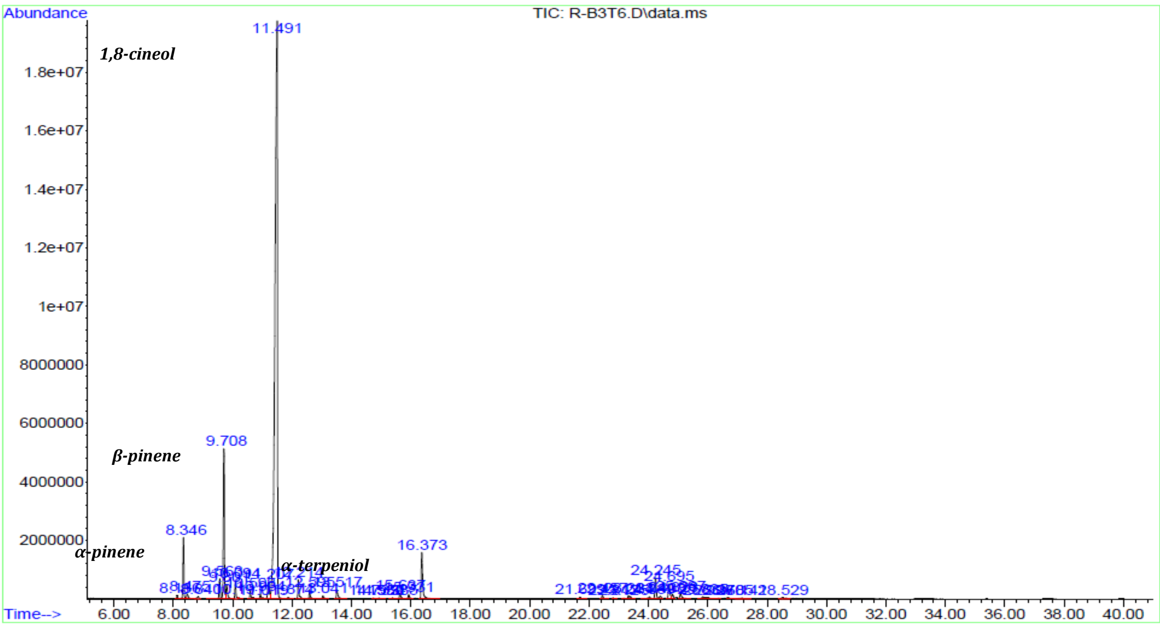

Fig. 7 The total gas chromatogram of the obtained cardamom essential oil

Table 2 Volatile compounds identified from essential oil on whole parts of Java cardamom

Essential oil	Formula	RT	Compound class	Percent
1,8-cineol	$C_{10}H_{18}O$	11.49	monoterpens	69.39
α-pinene	$C_{10}H_{16}$	9.7	monoterpens	9.24
β-pinene	$C_{10}H_{16}$	8.3	monoterpens	3.31
α-terpineol	$C_{10}H_{18}O$	16.4	Terpinene	3.56

Conclusions

The most important part of cardamom is the seeds yielding the highest essential oil. Under the selected parameters of the design of experiments, the maximum yield of cardamom essential oil was achieved with a cardamom fruit size of 9.52 mm and moisture content of 11% at 1 bar and 18 mL min^{-1} for 7h. Sixty-nine percent of cineole was contained in cardamom fruit and the rest are α-pinene, β-pinene, and α-terpineol. However, this does not mean that they are the best conditions to operate a distillation process applied to either cardamom seed or cardamom peel. There was no significant effect on essential oil yield when different durations were applied for the steam distillation method. Condensate flow rate is closely related to the steam flow rate which could affect the heat transferred toward the plant material and the steam pressure inside the distillation tank when it was being used. However, in this case, essential oil yield decreased when the condensate rate was higher than 18 mL min^{-1}. The possible reason was that the required dissipation of energy was not enough to extract the essential oil and non-heat resistance components could be easily degraded when exposed to large amounts of heat; thus, we concluded that process parameter should be set based on physical properties of material to obtain the economic essential oil.

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