Comparison of microwave hydrodistillation and solvent-free microwave extraction of essential oil from *Melaleuca leucadendra* Linn

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**Abstract.** The comparison of solvent-free microwave extraction (SFME) and microwave hydrodistillation (MHD) in the extraction of essential oil from *Melaleuca leucadendra* Linn. was examined. Dry cajuput leaves were used in this study. The purpose of this study is also to determine optimal condition (microwave power). The relative electric consumption of SFME and MHD methods are both showing 0,1627 kWh/g and 0,3279 kWh/g. The results showed that solvent-free microwave extraction methods able to reduce energy consumption and can be regarded as a green technique for extraction of cajuput oil.

1. Introduction

Essential oil, one of commodity export potential, could become a mainstay for Indonesia to earn foreign exchange. Essential oils are widely distilled in Indonesia, among others, patchouli, clove, nutmeg, lemongrass, vetiver, cajuput, and many others. Essential oils are the ingredients that are volatile (volatile), has a bitter taste, and smell like plant origin derived from plant parts such as leaves, fruit, seeds, flowers, roots, rhizomes, bark, even the whole plant. Furthermore, the development of essential oils are needed in the food flavoring industry, the cosmetics industry, and perfumery [1].

*Melaleuca leucadendra* Linn, commonly known as cajuput, is a plant growing at a level of 0 – 400 meter above sea level such as lowlands, along rivers, coastal areas or swamps. Cajuput can grow in barren, heat-resistant soil, and even sprout again after a fire. The spread of cajuput crops in Indonesia is found in several areas, namely Java, Irian Jaya, East Nusa Tenggara, Maluku, South Sumatra and Southeast Sulawesi. *Melaleuca leucadendra* Linn was first planted in Ponorogo, spread to Gunung Kidul in Yogyakarta, and other areas such as Gundih and Surakarta in Central Java, Mojokerto and Sukun in East Java, Cikampek, Majalengka, and Indramayu in West Java. Cajuput oil’s main compound is cineole (50-60%) with some other compound like alpha-terpineol, valeraldehyde, and benzaldehyde. This oil is used in herbal remedies such as analgetic, diaphoretic, antirheumatics, and spasmylytic [2]. The 1,8-cineole compound is the main compound of eucalyptus oil that acts as antimicrobial, antioxidant, immune, analgesic, and spasmylytic [3]. In addition, 1,8-cineole compounds also have the potential to be anti-inflammatory [4]. The methyl eugenol compounds and their derivatives, methyl isoeugenol are used in the perfume and fragrance industries [5].
The prospect of essential oil is relatively good in Indonesia, should have been followed by the mastery of refining technology and the cultivation of essential oil producing plants, so that businesses of essential oil is able to thrive in terms of quality and quantity to meet the world's needs and enhances competitiveness with suppliers from countries. The process of taking cajuput oil \((Melaleuca leucadendra\text{ Linn})\) is generally done by steam distillation and hydrodistillation. According to the previous research conducted by [2] using the steam-water hydrodistillation method on cajuput leaves \((Melaleuca leucadendra\text{ Linn.})\) delivered yields from 0.61 % to 0.85% of 5 hours of extraction time.

The previous research conducted by [6], namely the extraction of essential oil from orange peel takes 30 min using solvent-free microwave extraction while 180 min were required by hydrodistillation method. The optimum yield of essential oil obtained was 0.42 ± 0.02% by solvent-free microwave extraction and 0.39 ± 0.02% by hydrodistillation. It has shown the microwave extraction as an alternative method that can be developed more than conventional methods, due to high levels of product purity, minimal solvent usage, and short processing times. To optimize the process of essential oil extraction, this research will use microwave hydrodistillation and solvent-free microwave extraction method. The microwave hydrodistillation method is a combination of hydrodistillation and microwave use as heating [7]. The SFME method is a method of extracting essential oils without using a microwave-heated solvent which has advantages such as having a faster extraction rate, yield, and also higher purity of extract because it does not require solvent so it does not come into contact with chemicals [8].

2. Materials and methods

2.1. Materials

The \(Melaleuca leucadendra\) Linn leaf used in this study was collected from a forest owned by the Department of Forestry of Mojokerto (Fig. 1). All leaves were gathered from the same plantation age at approximately 0-2 years of plantation.

![Figure 1](image.png)

**Figure 1.** The location area where the \(Melaleuca leucadendra\) leaf was collected. The forest of the Department of Forestry is situated along Kemlagi-Mantup road and is easily accessed by any transportation vehicle.
2.2. Solvent-Free Microwave Extraction
The microwave oven used for solvent-free microwave extraction was Electrolux model EMM-2007X operating at 2450 MHz with a maximum delivered power of 600 W. The microwave oven was modified by drilling a hole at the top. Flat bottom flask having a capacity of 1000 mL was placed in the oven and connected to Liebig apparatus through the hole. After the Liebig condenser was placed into the oven, the hole around the neck of the flask was covered with polytetrafloraethylene (teflon) to prevent leakage of microwave.

For the extraction, sixty grams of dry Melaleuca leucadendra Linn. leaves were soaked in 700 mL distilled water at room temperature (25 °C) for an hour to hydrate the external layers of the plant material so that the excess water was drained off. The moistened plant material was placed in flat-bottom flask combined to a Liebig condenser. During the process, the vapor passed through the condenser outside the microwave cavity where it was condensed. The solvent-free microwave extraction process was performed for different mass and was continued until no essential oil obtained. The essential oil was collected, dried under anhydrous sodium sulphate and stored at 4 °C before used.

2.3. Electric Consumption
The electrical consumption of different extraction methods is calculated based on the effect of power consumption and time of extraction. The general equation for electricity consumption is explained by (Eq. (1))

\[ Ec = \frac{p \cdot t}{3600000} \]  

Where Ec is electric consumption (kWh), P is power consumption (W) and t is time (s). In addition, the relative electric consumption of the different methods of extraction can be expressed in equation 2.

\[ Ec* = \frac{Ec}{m*} \]  

Where Ec* is the relative electric consumption (kWh / g) and m* is the period of Melaleuca leucadendra Linn. (g).

2.4. CO\(_2\) emission
The CO\(_2\) calculation has been developed, according to the procedure mentioned in the previous study [6]. However, to get 1 kWh of energy from coal or fossil fuel, 800 g of CO\(_2\) will be released into the atmosphere during their combustion [6]. Then, the CO\(_2\) emissions are explained by (Eq. (3))

\[ E_{CO2} = \frac{Ec \cdot 800}{1000} \]  

Where \( E_{CO2} \) represents CO\(_2\) emission (kg).

For relative CO\(_2\) emissions different extraction methods are calculated according to (Eq. (4))

\[ E^{*} CO2 = \frac{E_{CO2}}{M*} \]  

Where E * CO\(_2\) is the relative CO\(_2\) emission (kg / g)
3. Results and discussion

3.1. Extraction yield of cajuput oil

The yield of cajuput oil was measured throughout the extraction process. Fig. 2 shows the variation of essential oil yield for solvent-free microwave extraction and microwave hydrodistillation processes at same power level. The figure shows both solvent-free microwave extraction and microwave hydrodistillation yields increasing as the mass of raw materials increase. The obtained results explain that solvent-free microwave extraction has more capability to generate cajuput oil rather than microwave hydrodistillation method and even the yields value is higher at the same extraction time.

![Figure 2. Extraction yield in cajuput oil extraction by solvent-free microwave extraction (SFME) and microwave hydrodistillation (MHD) methods](image)

It is known that the power extraction of microwave will control the amount of energy received by plant material, hence to be converted into heat energy. This heat energy helps the process of removal of essential oils from plant material or sample. The microwave power used in the extraction process by microwave hydrodistillation method is closely related to the process temperature, where the greater the power used, the system temperature at the extraction process will rapidly reach the boiling point of the used solvent. The power effect on the yield for the solvent-free microwave extraction method can be interpreted as the greater the microwave power used, the greater the energy received by the plant material to be converted into heat which eventually results in lower yields of cajuput oil. This case occurs because of thermal degradation of the material.

As depicted in Fig. 3, the more mass used, the weight of cajuput oil obtained decreases. However, the large amount of raw material and the amount of cajuput oil obtained, not always positively correlated with the increase in yield of cajuput oil obtained. This is because the yield of cajuput oil is influenced by the ratio factor of cajuput oil obtained and the mass of the initial raw material. In Fig. 3, there was a decrease in yield at a mass of 100 g, a decrease in the yield of cajuput oil by solvent-free microwave extraction method because the mass of the raw material (cajuput) used was excess (solid) and almost filled the pumpkin distiller. Where this results in a difficult vapor penetrating in the material to bring the molecular oil of the volatile oil diffuses out the material. The density of the material is closely related to the large space between materials. Excessively high and uneven density of materials can lead to the formation of steam lines that can reduce yields and quality of essential oils [9].
Figure 3. Extraction yield in cajuput oil extraction by solvent-free microwave extraction (SFME)

3.2. Electricity consumption and environmental considerations

The comparison of two different methods to extract cajuput oil with the same of both microwave power consumption and extraction time has been completed. The consumed energy to extract the cajuput oil with respect to the power consumption of the microwave is showed in Fig. 4. The relative electric consumption for generating 1 gram of cajuput oil using microwave hydrodistillation method is doubled the value of the solvent-free microwave extraction. The microwave hydrodistillation provide a value of 0.33 kWh/gram EO compared with 0.16 kWh/gram EO for solvent-free microwave extraction. This phenomenon suggests that the solvent-free microwave extraction is more efficient than the microwave hydrodistillation regarding the electric consumption.

The relative CO₂ emission produced of 1-gram cajuput oil is higher in the microwave hydrodistillation (0.13 kg CO₂/gram EO) than in the solvent-free microwave extraction (0.26 kg CO₂/gram EO) for an hour extraction time (Fig. 4).

Figure 4. Electricity consumption and environmental impact in cajuput oil extraction by solvent-free microwave extraction and microwave-assisted hydrodistillation methods
4. Conclusion
The present results demonstrate that the effect of the materials used the method of microwave hydrodistillation (MHD) and solvent-free microwave extraction (SFME) is the greater the material used, the greater the yield of cajuput oil produced. The optimum yield of essential oil was obtained 3.17 % by SFME and 1.93 % by MHD. In the extraction of cajuput oil, the solvent-free microwave extraction is more effective than the microwave hydrodistillation. Moreover, the solvent-free microwave extraction consumed less energy and less CO₂ emission than the microwave hydrodistillation did. Based on the variable of microwave power, the yield of cajuput oil produced less yield generated by solvent-free microwave extraction method when microwave power increased.

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