A COMPARISON OF ESSENTIAL OIL EXTRACTION FROM THE LEAVES OF LEMONGRASS (CYMBOPOGON NARDUS L.) USING TWO MICROWAVE-ASSISTED METHODS

Mahfud Mahfud1,*, Zuhdi Ma'sum1, Donny Satria Bhuana1, Ali Altway1, Yuyun Yuniati2

1Institut Teknologi Sepuluh Nopember
2Machung University

The extraction of essential oils from Lemongrass leaves (Cymbopogon nardus L.) has become more promising than ever before due to the oils' increasing demand in the market in addition to the dynamic breakthrough in its technology advancement. This study presents a comparison of two different methods of microwave-assisted essential oil extraction, namely Microwave Hydrodistillation (MHD) and Solvent-Free Microwave Extraction (SFME), both of which exhibit a better quality in terms of essential oil recovery compared to the conventional extraction methods. This study investigated the effect of extraction time, microwave power, as well as feed-to-distiller volume ratio (F/D). Experimental results suggest that although there is a tendency to increase yield along with the increasing power, the yield turns out to decrease at higher power, with the optimum power for the MHD method being 420 W and that of the SFME method being 560 W. Smaller F/D ratios appear to give higher yields for both methods and the smaller the size of the material, the higher the yield obtained. The MHD method produces a smaller yield but has a better quality of citronella oil compared to the SFME method. Two first-order and second-order extraction kinetics models were compared for both the MHD and SFME methods, and the results suggest that the first-order model was slightly better at representing the experimental data based on the RMSD and R² values. This applies to both experimental data using the MHD and SFME methods, respectively.

Key words: extraction, lemongrass oil, microwave hydrodistillation (MHD), solvent free microwave extraction (SFME)

INTRODUCTION

Located in a tropical climate, Indonesia is one of the countries that is geographically fertile for the diversity of plant species. Indonesian flora plays an important contribution to the world's total flora [1]. In addition, most plants in Indonesia provide benefits for human life, especially in the medical sphere [2] [3]. The production of essential oils is one form of plant empowerment to increase its selling value [4]. Basically, essential oils are already contained in the plant organs [5]. Essential oils are valuable because this type of oil is able to produce fragrances that contains volatile compounds [6]. The volatile compounds in essential oils have been studied to provide various benefits as antimicrobial, anti-inflammatory and aromatherapy [7] [8] [9]. Until now, essential oil production in Indonesia is continuously being developed and introduced to the world, by expanding the discovery of plant species, and finding effective and efficient methods. [10]. Lemongrass (Cymbopogon nardus L.) is a type of grass plant that has high prospects as an essential oil-producing commodity [11] [12]. Therefore, citronella is very suitable to be used as an object of current study related to the development of essential oil extraction methods [13]. Previously, the method of obtaining essential oils of a plant was carried out using the method of water distillation [14], and steam distillation [15] [16]. Unfortunately, some of these conventional extraction methods are considered less effective, especially in terms of the quality of the final product obtained [17]. The presence of heat energy is considered to affect the physical and essential oil characteristics, in which the chemical compounds in the final product are degraded [18] or mixed with solvent residues that are less green chemical [19]. This further emphasizes the reason for the development of effective methods for producing essential oils. Microwave-based extraction is a technology-based essential oil extraction method that is being developed because it is safe, without the use of harmful solvents and high thermal energy [20] [21]. In the future, this method will be used as a promising economic method for extracting the essential oil of a plant [22]. There are two types of extraction methods using microwave which are considered to be the most efficient in time and quality, namely Microwave Hydrodistillation (MHD) and Solvent-Free Microwave Extraction (SFME) [23] [24] [25]. The MHD method basically uses a combination of fast heating at microwaves with traditional solvent extraction [26], however, capable of producing high quality extract products only with a shorter extraction time, smaller amount of solvent, cheaper process and more environmentally friendly [27]. The SFME method promotes the simplicity in its working principle, which consists in the extraction of microwave-assisted dry distillation of a fresh plant matrix without adding any water or organic solvents and yet smell the best [28][29]. Comparisons were made by studying the effect of extraction time (t), microwave power (P) and feed to distiller volume ratio (F/D), as well as study of extraction kinetic models for these two methods.

*mahfud@chem-eng.its.ac.id
MATERIALS AND METHODS

Materials

In this study, the fresh lemongrass leaves used were obtained from the Pacet area, East Java, Indonesia. The initial treatment of materials, lemongrass leaves was stored in a dry place without sunlight for 2 weeks. The distilled water and anhydrous sodium sulphate used in the experimental work were of analytical grade.

Essential Oil Extraction

The mass of citronella leaves that have been weighed (40, 60, and 80 g) were put into a one-neck flask with a volume of 1000 ml. For the microwave hydrodistillation method, 200 ml of water was added, while for the solvent-free microwave extraction method, no water was needed. Heating process was carried out using microwave with the appropriate power variable (420, 560 and 700 Watt). The cooling process used a cold-water based condenser. In both methods, the water contained in the material and the water from the Clevenger apparatus to a distillation flask, which is known as cohabation, were both simultaneously recycled. It was done in such a way to prevent the material from burning faster and to avoid losing the oil that is still contained in the water distillate [30]. Extraction was carried out for 180 minutes with ten minutes interval for oil extraction. The percentage value of the yield of essential oil extract obtained was done by comparing the mass of oil extracted to the mass of the initial material, with the following equation:

\[
\text{Yield (\%)} = \frac{\text{mass of oil extracted}}{\text{mass of the initial material}} \times 100\%
\]  

Kinetic Models of Extraction

Two kinetics models examined are 1st-order and 2nd-order models for Microwave Hydrodistillation (MHD) method as well as the Solvent Free Microwave Extraction (SFME) method. The 1st-order kinetic extraction equation used from the previous work [31] [32] can be written in the following differential form:

\[
\frac{dC_t}{dt} = k_1 (C_S - C_t)
\]

\[
C_t = C_S (1 - e^{-k_1 t})
\]

Where: \( k_1 \) (min\(^{-1}\)) is the extraction rate constant for 1st-order model, \( t \) (min) is time of extraction process, \( C_t \) is the extraction yield (%w/w) at \( t \), \( C_s \) is the extraction yield maximum or extraction capacity. Hence the boundary conditions are \( C_t = 0 \) at \( t = 0 \) and \( C_t = C_s \) at \( t = \infty \). Upon solving the ODE, Eq. (2) reduces to Eq. (3). The 2nd-order rate law, where the effect of microwave heating that can break-out of matrix material containing oil into a solution can be expressed in a second-order model. The extraction rate equation of can be written as follows:

\[
\frac{dC_t}{dt} = k_2 (C_S - C_t)^2
\]

\[
C_t = \frac{C_S^2 k_2 t}{t + C_s k_2 t}
\]

Where \( k_2 \) (L g\(^{-1}\) min\(^{-1}\)) is the extraction rate constant for 2nd-order model, \( C_s \) is the extraction yield maximum or extraction capacity. And the boundary conditions are \( C_t = 0 \) at \( t = 0 \) and \( C_t = C_s \) at \( t = \infty \). Therefore, the integration of Eq. (4) become an equation (5) in the form of \( C_t \) function of time of time. The model parameter \( k_1 \) and \( k_2 \) model parameters were obtained by fitting the parameter method between the experimental data and the model calculation results through equation (3) by minimizing root-mean-square deviation (RMSD) using the solver application contained in Microsoft Excel and adjusted correlation coefficient (R2 adj) also evaluated. The low value of RMSD and a high R2 adj value indicate a good fitting between experimental data and the models [33] [34]. The values of RMSD and R2 are calculated using equation (6) and equation (7):

\[
\text{RMSD} = \sqrt{\frac{1}{n} \sum_{i=1}^{n} (C_{\text{calc},i} - C_{\text{exp},i})^2}
\]

\[
R^2 = 1 - \frac{(n - 1) \cdot \sum_{i=1}^{n} (C_{\text{calc},i} - C_{\text{exp},i})^2}{(n - 1 - m) \cdot \sum_{i=1}^{n} (C_{\text{exp},i} - C_{\text{exp},i})^2}
\]

where \( C_{\text{calc}} \) and \( C_{\text{exp}} \) are calculated by models and experimental yields of oil extracted, \( C_{\text{exp}} \) is experimental data average. Then \( n \) is the number of experimental data and \( m \) is number of independent variable.

RESULTS AND DISCUSSIONS

The Extraction Time effect on Citronella Oil Yield

The extraction time is one of the factors that need to be considered in the extraction of citronella oil. Figure 1 shows that the quantity of citronella essential oil obtained by the SFME method is larger than that from the MHD method. The percentage of yield using the SFME and MHD methods was 1.45% and 1.36%, respectively. In terms of appearance, citronella essential oil produced using the MHD method has a clearer appearance compared to the SFME method [35]. The extraction of citronella oil with the method of microwave hydrodistillation and Solvent-Free Microwave Extraction show that the citronella oil obtained increases rapidly at first and then remains constant afterwards with the extraction time.
Based on the experimental results, the effective extraction of citronella essential oil was carried out for 150 minutes (Figure 1). In general, the longer the extraction time, the higher the yield obtained, following an exponential pattern [36]. This is proven by the extraction process using the MHD and SFME methods, as shown in Figure 1.

![Figure 1: The effect of the extraction time on yield for MHD and SFME. (P = 420W, F/D = 60 gr/L)](image)

The extraction process for citronella essential oil can be divided into 3 extraction phases, namely: 1) equilibrium phase (0-60 min), 2) transition phase (60 min) and 3) the diffusion phase (140-180 min). The equilibrium phase is the phase that occurs at the beginning of the extraction process, where there is a sharp increase in yield which is directly proportional to the length of the extraction time. The transition phase is a phase of increasing the yield value which tends to decrease slightly when compared to the equilibrium phase along with the addition of extraction time. The diffusion phase or what can be called the limiting phase is the phase where very little oil is extracted, so that the recovery of essential oils tends to be constant. During the diffusion phase, the extraction was stopped because the addition of extraction time was considered to have less significant effect on the yield [37]. Therefore, 180 minutes is the most effective time to extract essential oils, both using the MHD method (1.2% yield) and SFME (1.4% yield).

**The microwave power effect on citronella oil yield**

The microwave power plays an important role in controlling the amount of electromagnetic wave transferred to the media in the distiller to be converted into heat energy. This heat energy will cause the expansion of the oil, then break plant matrix that protects the essential oil, so that the essential oil comes out and is easily extracted. The presence of high microwave power has been shown to accelerate the hydrolysis of the extracted material [38]. The effect of microwave power on yield of citronella oil can be seen in Figure 2. In general, the use of the SFME method resulted in slightly higher yields than the MHD method at the same power. Paying attention to the experimental results curve gives an idea of how there is a tendency to increase the yield when the power is increased. It is said that the greater the energy, the higher the yield value.

![Figure 2: The effect of microwave power to extraction on yield for MHD and SFME](image)

However, at a certain turning point or a certain phase, when the power is increased continuously, the resulting yield actually decreases. The turning point in the MHD method occurs when the microwave power is 420 W, while the SFME method is 560 W. Therefore, the turning point is used as the optimum condition for the use of microwave power, resulting in a yield percentage of 1.35% (MHD, 420 W), and 1.59% (SFME, 1.6%).

**The feed to distiller volume (F/D) effect on citronella oil yield**

The difference in the feed-to-distiller volume ratio (F/D) also affects to the oil obtained, as shown in Figure 3. The increase in the feed-to-distiller volume ratio will decrease the extraction yield. The greater the mass of the material, the smaller the cavity, the density of the mass of the material and the surface area of the material. This factor will reduce the ability of essential oil extraction to get out of plant organ tissue properly.

![Figure 3: The effect of feed to distiller volume ratio (F/D) on oil yield for MHD and SFME](image)

This is the reason why the yield tends to decrease as the mass of the extracted material increases [39] [40]. The mass of lemongrass leaves is 20 g, or equivalent to the ratio of feed intake to volume of water solvent (F/S) for MHD 0.1 g/ml and distiller volume (F/D) for SFME of 0.02 g/ml is the weight of the raw material.
used. It is recommended to extract essential oils in order to obtain the optimum yield, namely the percentage yield of 1.34% (MHD) and 1.53% (SFME).

**Kinetic Modeling for Oil Extraction with MHD and SFME**

In both methods using MHD and SFME, the yield of citronella oil increased rapidly at first along with the extraction time but then there was a slight increase and finally there was no further increase. The yield of citronella oil as a function of time is shown in Figure 1. This trend is commonly found in the literature for both MHD [24] [26] [27] and SFME methods [25][28]. This can be used to determine the kinetic model for both oil extraction methods. In the extraction of citronella oil by the MHD method, there are two mechanisms. The first mechanism is as in the conventional essential oil distillation process, namely water as a solvent will absorb the waves so that it becomes hot, then heats up the leaf matrix so that the oil comes out by diffusion and is carried away by water. The second mechanism is the essential oil in the plant matrix containing organic compounds that can absorb electromagnetic waves so that heating occurs in the oil and changes in the matrix cell structure caused by electromagnetic waves. In this case, the microwaves interact with water molecules present in the vascular matrix system as well as with water molecules as solvent. Thus the oil expands dramatically, which is followed by tissue rupture. The rupture of this tissue then allows the volatile oil components to escape. Organic compounds contained in essential oils have high or low dipolar moments. Organic compounds that have a high dipolar moment will interact more easily with microwaves and can be extracted more easily compared to organic compounds that have a low dipolar moment. While the extraction of citronella oil with the SFME method does not use solvent or water, the presence of water is only found in the matrix of the material whose amount corresponds to the water content of the material. The mechanism that occurs is almost the same as the MHD method, however, since water does not present as a solvent, microwaves are only absorbed by water and oil in the material so that there is no loss of energy absorbed by solvents outside the material, thus, making this process more energy efficient. The release of oil depends only on the speed with which the oil component absorbs the energy from the microwaves and the water contained in the material matrix. Therefore, the phenomenon of microwave-assisted extraction is not purely an event of mass transfer or dissolution of the material into the solvent, which in general can always be expressed by an extraction model of order 1. Many other authors have developed several models of microwave-assisted extraction, including Empirical Kinetic Models, Power Law Models, Hyperbolic Models, Weibull’s exponential equation and Elovich's logarithmic equation [35]. However, considering the complexity of the microwave-assisted extraction phenomenon, most authors use a simple model, namely the simple model of order 1 [16] [41] [42] and order model 2 [43-46]. Hence in this study, both experiments with the microwave hydrodistillation (MHD) method and experiments with the solvent free microwave extraction (SFME) method used simple 1st-order and 2nd-order models based on equation 2 and equation 4. The results of the comparison of model 1 and model 2 of the microwave hydrodistillation (MHD) method are shown in Figure 4 and for the solvent free microwave extraction (SFME) method are shown in Figure 5.

![Figure 4: Comparison two kinetic models with the experimental data for MHD (420 W, 2 cm, 60 g/mL)](image-url)
to distinguish. However, Table 1 shows that the first-order model has a lower RMSE and a relatively higher coefficient of determination (R2). Thus, it can be said that the first-order kinetic model represents the results of the citronella oil extraction experiment better than the second-order, both for the MHD and SFME methods.

**CONCLUSIONS**

The extraction of citronella oil with the method of microwave hydrodistillation and Solvent-Free Microwave Extraction show that the citronella oil obtained increases rapidly at first and then remains constant afterwards with the extraction time. Based on the experimental results it was found that the effective extraction of citronella essential oil was carried out for 150 minutes. So far, the SFME method is considered to provide a slightly higher yield than the MHD method. The effect of microwave power on the yield of citronella essential oil shows that the SFME method produces a slightly higher yield than the MHD method, for the same microwave power and there is a tendency to increase yield along with increasing power, meaning that the greater the energy used in extraction, the higher the yield. The difference in the ratio of ingredients and the volume of distiller (F/D) of citronella samples also affects the percentage of yield obtained, the yield tends to increase with the ratio of ingredients and volume of the distiller. The more the mass of the material increases, the smaller the hollow density of the material and the surface area of the material, thus weakening the ability of essential oil extraction to get out of plant organ tissues. This causes the yield to decrease as the mass of the extracted material increases.

In the kinetic modeling of citronella oil extraction, the first-order kinetics models for extraction by microwave hydrodistillation (MHD) and solvent-free microwave extraction (SFME) methods represent the experimental results of citronella oil extraction better than second order.

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