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Intensification of Kalimantan White Pepper (Piper nigrum) Oil Extraction Based on Microwaves and Ultrasonics

Intensifikasi Ekstraksi Minyak Lada Putih (Piper Nigrum) Kalimantan Berbasis Gelombang Mikro dan Ultrasonik

Bangkit Gotama*, Khalisa Addina, Nur Afrie Rizqie Ananda

Department of Chemical Engineering, Institut Teknologi Kalimantan

*Corresponding author: bangkit.gotama@lecturer.itk.ac.id

ABSTRACT

White pepper is one of the staple spices of East Kalimantan. In an attempt to boost the competitiveness of white pepper commodities, it is necessary to raise the added value of the commodity by transforming it into high-value goods, such as white pepper oil. This research intends to evaluate the production of local (LPL) white pepper oil (MLP) with commercial white pepper oil (LPK) using different extraction techniques (microwave-assisted hydro distillation, MAHD; and ultrasonic following hydro distillation, US-HD. This experiment also evaluated the effect of extraction factors on oil yield, including microwave power on MAHD, ultrasonic wave power on US-HD, sonication duration on US-HD, and solvent volume. A comparison of the energy usage of both approaches was also conducted in an attempt identify the most effective approach. The MAHD and US-HD techniques were used to extract 80 grams of dry white pepper powder mixed with water for 90 and 180 minutes, respectively. Specifically for US-HD, the combination of raw materials was sonicated for 3-5 minutes before to extraction utilizing the HD method. In both MAHD and US-HD, LPK includes more MLP than LPL, according to experimental findings. MLP yield may be increased by the addition of solvent, wave power (micro and ultrasonic), and sonication time. Based on its efficiency and energy consumption, the US-HD approach is superior to the MAHD.

Keywords: *white pepper, essential oil, extraction, microwave, ultrasonic*

ABSTRAK

Lada putih merupakan salah satu rempah andalan Kalimantan Timur. Dalam upaya meningkatkan daya saing komoditas lada putih, perlu langkah strategis untuk meningkatkan nilai tambah komoditas menjadi produk bernilai tinggi, yakni minyak lada putih. Penelitian ini bertujuan untuk membandingkan produk minyak lada putih (MLP) lokal (LPL) dan komersial (LPK) dengan berbagai metode ekstraksi (*microwave-assisted hydro distillation*, MAHD; dan *ultrasonic following hydro distillation*, US-HD. Di samping itu, evaluasi parameter ekstraksi terhadap rendemen minyak seperti daya gelombang mikro pada MAHD, daya gelombang ultrasonik pada US-HD, waktu sonikasi pada US-HD, dan volume pelarut turut dikaji di dalam eksperimen ini. Analisis konsumsi energi pada kedua metode tersebut turut dilakukan untuk memperoleh metode yang lebih efisien. 80 gr bubuk lada putih kering yang dicampur dengan aquadest diekstraksi baik dengan metode MAHD maupun US-HD, masing-masing selama 90 dan 180 menit. Khusus pada US-HD, campuran bahan baku disonikasi terlebih dahulu selama 3-5 menit sebelum diekstraksi menggunakan prosedur HD. Berdasarkan hasil eksperimen, LPK mengandung MLP yang lebih tinggi daripada LPL baik pada MAHD maupun US-HD. Pertambahan pelarut dan daya gelombang (mikro dan ultrasonik); serta waktu sonikasi sama-sama dapat meningkatkan MLP. Menurut evaluasi produktivitas dan konsumsi energi, metode US-HD lebih unggul dibandingkan dengan MAHD.

Kata kunci: lada putih, minyak atsiri, ekstraksi, gelombang mikro, ultrasonik

INTRODUCTION

Pepper, or King of Spice, also referred to as pepper, is a spice valued for its advantages, particularly as a culinary spice and raw material for the food industry. In addition, pepper may be used as an insecticide, herbal medication containing anti-fungal, anti-bacterial, and anti-bacterial components, and in the cosmetics business (Wang et al., 2018). Moreover, both white pepper and black pepper are rich in bioactive substances such as piperine, flavonoids, polyphenols, and essential oils. In particular, pepper essential oil includes aroma-and flavor-contributing compounds such as α-pinene, β-pinene, linalool, phellandrene, limonene, myrcene, caryophyllene, and α-copaene. Similar to pepper, pepper oil has antioxidant, analgesic, antibacterial, and insecticidal properties (Elfahdi, 2021).

Pepper is one of Indonesia's most important spice crops, particularly in East Kalimantan. Indonesia is one of the world's largest producers of pepper. According to Ministry of Trade statistics, Indonesia was the third biggest exporter of pepper in the world in 2019, after Vietnam and Brazil, with an 8.66% market share (Sampepana et al., 2020). The following year, in 2020, exports of white pepper reached 24.788 tons with a value of $75.316,2 (Badan Pusat Statistik, 2021). The high export value led to a high export volume, which reached 3.760 tons in 2020 (BPS Kalimantan Timur, 2022). However, pepper production in East Kalimantan has not increased the competitiveness of pepper commodities on a national or worldwide scale. Due to the poor quality of pepper, a number of strategic initiatives are required to enhance product quality and increase the commodity's competitiveness (Sampepana et al., 2020). Using good extraction technology, white pepper products could be turned into essential oils. This would make the pepper industry more competitive.

Hydro distillation (HD), steam distillation (SD), and solvent extraction (SE) (Tran et al., 2019; Tran et al., 2020), as well as innovative non-conventional ones such as microwave-assisted hydro distillation (MAHD), microwave-assisted extraction (MAE), and ultrasound-assisted extraction (UAE) are used for the extraction of pepper oil. Conventional techniques have several shortcomings, including lengthy extraction periods; significant energy use; and poor oil yield. On the other hand, non-conventional technologies are able to overcome these limitations, making their industrial use highly promising (Wang et al., 2018). However, it is still difficult to obtain experimental research on the extraction of white pepper oil in East Kalimantan, particularly those conducted by indigenous farmers who use non-conventional extraction techniques. This is why this study must be conducted immediately.

This research intends to compare the yields of local white pepper (LPL) and commercial white pepper (LPK) to those of white pepper oil (MLP) when extracted using different techniques. In addition, this experiment assessed the effect of extraction factors on oil yield, including microwave power on MAHD, ultrasonic power on US-HD, sonication duration on US-HD, and solvent volume. In this research, the amount of energy used by each method was also looked at to find the best way to do things.

MATERIALS AND METHODS

Materials

This study used both indigenous and commercial white pepper. Pepper producers in Semoi Dua Village, Sepaku District, North Penajam Paser Regency, East Kalimantan provided the local white pepper (LPL). The obtained LPL was then sun-dried till its moisture content reached 4.5%. In addition, LPL was crushed and sieved through a 30-mesh sieve before being kept at room temperature in a Ziploc bag before extraction. In this investigation, powdered commercial white pepper (LPK) of the Koepoe-Koepoe brand (PT. Anggana Catur Prima, Jakarta) was used for comparison of raw materials prior to extraction. As the solvent for this study, PT Jayamas Medica Industri, Sidoarjo, used distilled water with the brand name WaterOne.

Equipment

MAHD and US-HD are two different ways of doing things, so the experimental equipment used in this work is also different. Figure 1 shows that the main tool for MAHD is a microwave oven (Samsung ME731K), while Figure 2 shows that the main tools for US-HD are an ultrasonic cleaning bath (Elmasonic P60 H) and a heating mantle (Electrothermal EM0500 /CE) as a heating source.

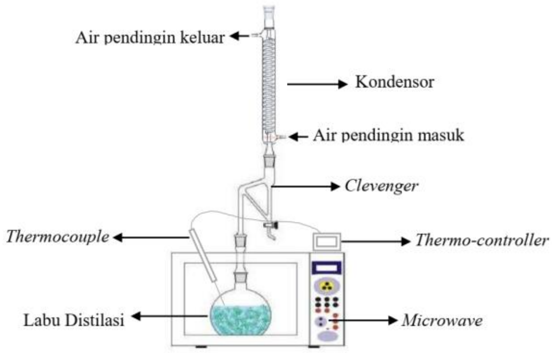

Figure 1. Schematic of the equipment used to study the extraction of white pepper (LPL/LPK) using the MAHD method.

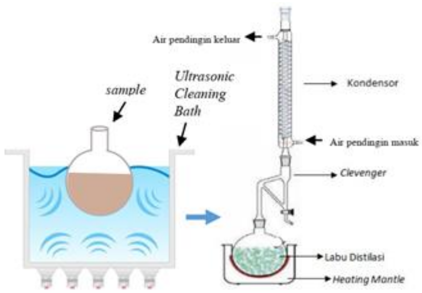

Figure 2. Schematic of experimental apparatus for white pepper extraction study (LPL/LPK) using the US-HD technique.

Methods

This is the first time the MAHD technique has been used to extract white pepper oil (MLP). In the extraction flask, 80 grams of white pepper powder, LPL, and LPK were weighed and combined with distilled water based on the variables (320 and 400 ml). Next, the microwave oven's power level is adjusted based on the specified factors (300 and 450 watts). The extraction procedure was then conducted for 90 minutes, determined from the moment the distillate started to drip from the condenser. Following the completion of the extraction procedure, the oil is removed from the hydrosol solution and combined with sodium sulfate to absorb the leftover water. Next, the mass of the oil is determined so that the MLP yield can be estimated.

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The second approach, US-HD, starts with the weighing and mixing of basic components, similar to the first method. The pepper powder and solvent combination in the extraction flask were sonicated for 3-5 minutes in an ultrasonic cleaning bath. The used ultrasonic power ranges from 135 to 150 watts. The combination of pepper powder and water was then extracted by hydro distillation (HD) using a heated mantle. This HD operation lasted 180 minutes, commencing when the distillate began to trickle. After the HD distillation process was done, the next steps were done in a way that was similar to the MAHD process.

RESULTS AND DISCUSSION

Comparison of White Pepper Raw Material Productivity

To determine the potential of local white pepper for manufacturing high-value white pepper diversification products, specifically MLP utilizing the MAHD technique, a comparison of white pepper raw materials (LPL and LPK) must be conducted. LPL is derived from the modest output of local farmers. LPK is derived from pepper seeds that have undergone more advanced processing (spice industry) than LPL. In general, as shown in Figure 3, the experimental findings indicate that LPL contains fewer essential oils than LPK. The most MPL came from LPK extraction with 450 watts of power, while the least came from LPL extraction with 300 watts of power.

In addition to the MAHD approach, US-HD was also used for the comparative assessment of white pepper raw materials. According to Figure 4, MLP in LPL is comparable to LPK at 135 watts and 150 watts. In LPK extraction, the maximum and minimum MLP are obtained. On the other hand, LPL has more MLP than LPK at lower power, while LPK is better at greater power.

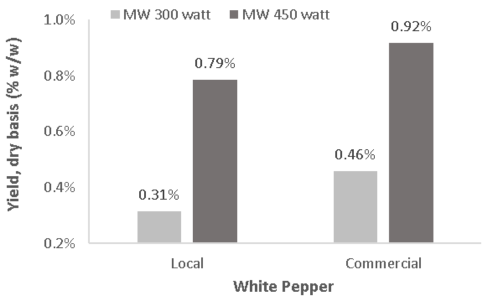

Figure 3. Variations in MLP for different white pepper raw material sources and microwave power utilizing the MAHD technique.

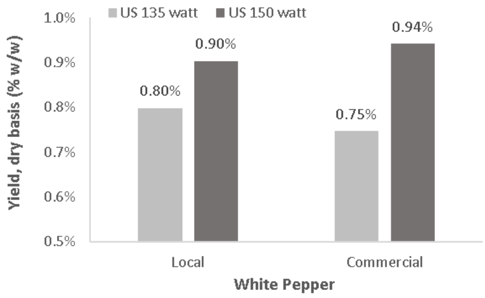

Figure 4. Variations in MLP for different white pepper raw material sources and ultrasonic wave power with the US-HD technique.

Effects of Solvent Volume and Wave Energy

Parameters like solvent volume and wave power, both micro and ultrasonic, play a vital role in MLP extraction. In MAHD, microwave irradiation accelerates plant cell death, followed by a fast diffusion rate of intracellular components (oils) into the solvent (Chemat et al., 2020). Similarly, in US-HD, the strength of ultrasonic waves may induce the development of microbubbles that can break plant cell walls, resulting in a simultaneous increase in the rate of solvent penetration into cells and the rate of mass transfer (Yu et al., 2021). Water is the only environmentally friendly solvent that is simple to apply to both MAHD and US-HD. This is due to the high value of water's dielectric constant, which facilitates

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the solvent's ability to absorb energy, allowing it to readily raise the temperature of the raw material combination and enable the oil's release (Lefebvre et al., 2021).

The profile of MLP on LPL extraction using the MAHD approach is shown in Figure 5. The graph demonstrates that the quantity of solvent has a favorable effect on MLP growth. This is consistent with what Chen et al. (2020) indicated about deulkkae oil extraction, namely that with the right quantity of solvent, the substance and solvent would interact enough to lower mass transfer resistance and facilitate the release of oil from plant cells. In addition, the increase in power has a positive effect on MLP. Drinić et al. (2021) reported that with higher power extraction of ironwort oil, they were able to create a higher extraction temperature, where extraction efficiency was strongly influenced by system temperature and the stability of the target component. However, exceeding the optimal power level might have a detrimental effect on oil production.

At the highest microwave power (450 watts), there was a considerable improvement in yield (57.9%) owing to an increase in solvent volume (from 320 ml to 400 ml). While at a lesser power, at 300 watts, the results do not increase by 10%, they do increase by 5%. In relation to the increase in microwave power (from 300 to 450 watts), the MLP yield increased to 150% at a solvent capacity of 400 ml, but only achieved 72.7% at a lower volume (320 ml). So, we can say that 450 watts of power and 400 ml of solvent work well together for LPL extraction using the MAHD method.

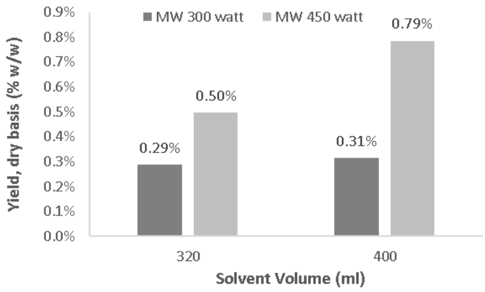

Figure 5. Effect of solvent volume and microwave power on the MAHD extraction of MLP on LPL.

On the US-HD approach, an analysis of MLP enhancement was also conducted, with almost comparable MLP patterns. Figure 6 illustrates the rise in MLP during LPL extraction with the US-HD technique, where both the addition of solvent and ultrasonic power result in an increase in MLP. These findings are consistent with the cinnamon oil extraction experiments conducted by Chen et al. (2021), namely increasing the irradiation power of ultrasonic waves to optimal conditions capable of destroying cell tissue and accelerating the mass transfer rate of oil components into the solvent. Also, the research showed that with an ideal volume of solvent, microwave adsorption on materials can be increased to give enough energy to help break down cell walls and let oil out (Chen et al., 2021).

In general, the MLP is greater with a solvent capacity of 400 ml compared to 320 ml. The same occurs with ultrasonic wave power, which is 150 watts more yield than 135 watts. Increases in solvent volume (from 320 to 400 ml) led to a 64% increase in yield. However, adjustments in wave power (from 135 to 150 watts) were unable to boost MLP by more than 14%. Therefore, it can be inferred that yield modifications may be optimized by increasing the solvent volume.

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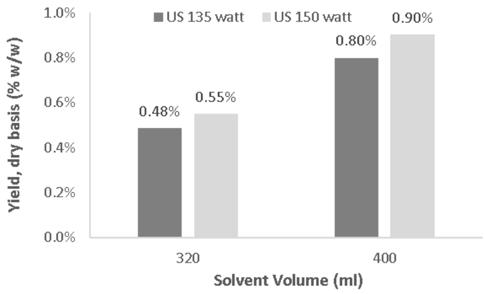

Figure 6. Effect of solvent volume and ultrasonic wave power on MLP on LPL extraction using the US-HD method.

Time Sonication's Influence on US-HD

In addition to the wave power and solvent volume, the sonication period has a major effect on the MLP in US-HD. According to Yu et al. (2017), the longer the duration of sonication, the more intensely the generation of collapsing bubbles can damage the cell walls of the plant matrix, the simpler it is for the solvent to extract the oil.

According to Figure 7, the length of sonication is directly related to yield at both 135 and 150 watts, which is consistent with the findings of Chen et al. (2021). 5 minutes of sonication at 150 watts produced the greatest MLP, while 3 minutes of sonication at 135 watts produced the least yield, 0.45%. At 150 watts of power, the largest yield acceleration (63.9%) occurs between 3 and 4 minutes, after which the speed begins to decelerate. This is in stark contrast to the 135 watt extraction, which witnessed the greatest yield acceleration (38.6%) between 4 and 5 minutes.

Extraction Method Comparison

To determine whether the MLP extraction process is preferable between MAHD and US-HD, it is required to compare oil productivity and energy consumption per mass of MLP. Both of these approaches are cutting-edge extraction technologies that may boost quality and output while overcoming the limitations of conventional extraction techniques (Radivojac et al., 2021). However, MAHD and US-HD are both able to boost extraction efficiency; their methods and capacities are distinct.

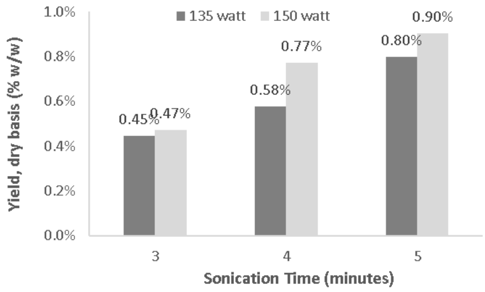

Figure 7. Effect of sonication time on MLP on LPL extraction using the US-HD method.

Figure 8 demonstrates that there is a difference in yield between the MAHD and US-HD procedures based on their respective extraction yields. Based on the acquired findings, the US-HD approach is more productive than the MAHD methods employing LPL or LPK. This phenomena relates to the research done by Chen et al. (2020) , in which the percentage discrepancy between the findings exceeded 30%. In particular, for the extraction of MLP from LPL raw material, the US-HD yield is 15% higher than that of the MAHD. In contrast, there was no significant difference between the MAHD and US-HD approaches for the extraction of MLP from LPK (percentage difference less than 3%).

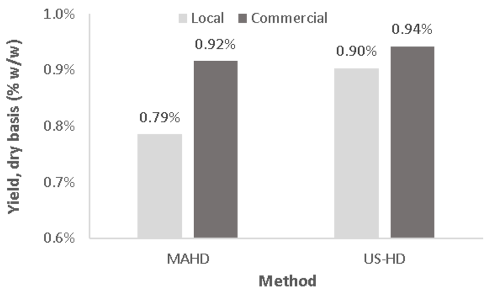

Figure 8. Comparison of the MAHD and US-HD Extraction Techniques for MLP on LPL.

Even though the US-HD approach takes a longer extraction time, particularly with

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the 180-minute conventional heating procedure after the sonication step, the energy consumption per unit mass of the MLP product is much lower than that of the MAHD method, for both LPL and LPK. This is a result of the comparatively low energy consumption of the heating mantle and the increased productivity of US-HD compared to MAHD (Figure 9). The restriction of maximum power utilization on microwave oven equipment to 450 watts (to prevent entrainment phenomena) is a significant factor in the energy consumption of MAHD. In reality, it takes more than twice as much energy to run the instrument as it puts into the extraction flask.

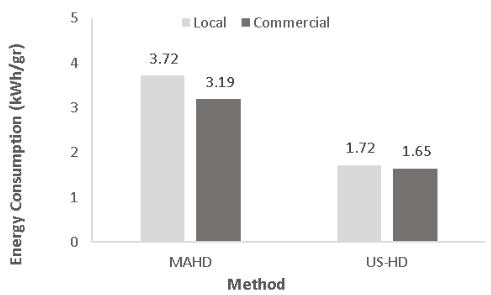

Figure 9. Comparison of the energy consumption of the MAHD and US-HD techniques for LPL extraction.

The yield difference between the two approaches falls between 1.5 and 2.0 percentage points. US-HD is a very dependable approach in a variety of experimental situations when yield differences between raw materials do not exceed 5%. Even though the MAHD method tends to be less accurate, when the difference is more than 16%. Table 1 is used as an extra source of data in the calculation process so that the energy consumption can be studied.

Table 1. Specifications of data extraction equipment for the estimation of MLP extraction energy consumption.

Equipment Specification	Microwave Oven[a]	Ultrasonic Cleaning Bath[b]	Heating Mantle[b]	Chiller	Cooling Water Pump
Power input (watt)	1.150	580	200	100	25
Extraction time (minutes)	90	3-5	180	90[a] and 180[b]	90[a] and 180[b]
Heating time (minutes)	15	-	30	15[a] and 30[b]	15[a] and 30[b]

[a] MAHD
[b] US-HD

CONCLUSION

On average, all extraction techniques revealed that LPK contained 0.46–0.94% more essential oil than LPL (0.31–0.94%).

450 watts of microwave power, 90 minutes of extraction, 80 g of pepper, and 400 ml of solvent yielded the greatest MLP content in MAHD (0.79 % LPL and 0.92 % LPK). On the other hand, the highest yield of US-HD (0.90 % LPL and 0.94 % LPK) was achieved with a power of 150 watts, sonication for 5 minutes, and extraction for 180 minutes. The ingredients were 80 grams of pepper and 400 ml of solvent.

In addition, US-HD is superior to MAHD since it requires 50% less energy to extract the same quantity of oil.

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