Sonication Effect on Modified Ultrasound Assisted Aqueous Extraction (UAAE) of Rice Bran Oil Yield

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Abstract. Rice bran oil (RBO) is extracted using ultrasound-assisted aqueous extraction (UAAE) with minor modifications to the separation process. Although conventional methods such as solvent extraction or mechanical pressing are more commonly used in the RBO industry, UAAE is one of intensification processes that can increase RBO productivity. In this study, sonication operating conditions such as time and temperature and solid/liquid to ratio were studied to obtain the optimum yield. The results indicated that the sonication time and temperature positively affected the yield increase, while SLR led to a decline after reaching 0.2 g/ml. The highest yield was obtained at the sonication time of 60 seconds, SLR 0.2 g/ml, and sonication temperature of 40°C, 17.6%. Besides, the relative energy consumption of UAAE at temperatures of 20 and 40°C, and aqueous extraction (AE) were also investigated. Besides producing the highest yield, UAAE at 40°C has a lower relative energy consumption than others, so it can be considered a prospective method of RBO extraction. Further development on UAAE and its combination with other methods are needed to fulfill economies of scale of the RBO industry.

Keywords: Rice bran oil, sonication, ultrasound-assisted aqueous extraction, yield.

1. Introduction
Rice bran is one of the byproducts collecting from milling rice grain into rice other than a husk. Approximately 10% of the grain weight is rice bran [1]. In 2019, Indonesian grain production (gabah kering giling or GKG) reached 54.6 million tons or approximated 5.46 million tons of rice bran. Whereas in North Penajam Paser Regency, East Kalimantan Province, grain production reached 41,622 tons, or around 4,162 tons of rice bran was obtained from milling [2]. However, the amount of rice bran production is not proportional to its utilization. To date, rice bran has only been used as animal feed, particularly in the Babulu Subdistrict of Penajam Paser Utara Regency. Rice bran is a potential food ingredient with numerous nutraceutical content such as tocopherols, phytosterols, squalene, γ-oryzanol, and polyphenols [3]. In addition, rice bran also contains 16-19% fat; 12-14% protein; 41-45% carbohydrate; 12-13% fiber; 10-11% ash; and the rest are minor components [4]. Rice bran oil (RBO) obtained from rice bran extraction using the solvent extraction consists of saturated fatty acid, monounsaturated fatty acids, polyunsaturated fatty acid, and unsaponifiable matter [5]. Some popular extraction methods frequently used in RBO extraction are cold pressing [6]–[10], aqueous extraction (AE) [1] [11][12], solvent extraction [3], [13]–[15], and soxhlet extraction [16]. Nowadays, non-conventional methods are popular because they can produce higher productivity than conventional methods; one of them is the ultrasound-assisted extraction (UAE) based on both organic and water solvents [17], [18]. The method utilizes ultrasonic wave radiation (sonication) to increase the extraction process’s effectiveness by intensifying mass and heat transfer between the solvent and...
the target component [19]. Sonication to the solvent extraction can enhance oil yield by 29.34% [13] as well as to the soxhlet extraction by 71.16% [16]. Also, the combination of supercritical fluid extraction with sonication can increase yields up to 27% compared to those without sonication [20]. Additionally, many authors have developed non-conventional methods consist of microwave-assisted extraction or MAE [21], [22] and extraction with supercritical fluid extraction (SFE) [20].

RBO extraction with the UAE method usually uses organic solvents such as n-hexane, ethanol, methanol, and ethyl acetate. The solvents are less environmentally friendly and healthy [19] if the process of separation between the solvent and the RBO is not perfect. Therefore, green solvents such as water in RBO extraction need to be developed. Besides being inexpensive, water is harmless and environmentally friendly [23]. Some authors used water on RBO extraction using aqueous extraction [11], [12], [18]. Khoei and Chekin [18] studied RBO extraction with ultrasound-assisted aqueous extraction or UAAE combining the sonication step with aqueous extraction. Nevertheless, the process carried out takes a long time. It was caused by the duration of the freezing of water-oil emulsion. In our research, we develop a new procedure replacing the freezing step in the previous method with the evaporation process to separate oil from the solvent. Therefore, this study investigates the influence of sonication parameters of modified UAAE on productivity and relative energy consumption. The results are expected to develop the RBO extraction process design improving utilization of rice milling waste.

2. The Material and Method

2.1. Sample and chemical.
Rice bran was provided by local farmers in Babulu District, Penajam Paser Utara Regency, East Kalimantan Province. Before using it, the bran was stabilized by heating in an oven at 110°C for 20 minutes to overcome the bran's enzymatic reaction [13]. Then the bran is filtered using a 30-mesh filter [24] and stored in a plastic bag. Aquadest purchased from local producers in Balikpapan, East Kalimantan.

2.2. Sonication.
From the previous step, rice bran was weighed and mixed with aquadest as much as 100 ml in a beaker glass to achieve a solid/liquid ratio (SLR) of 0.1 to 0.3 gr/ml. After that, sonication was performed with a horn sonicator with 350 watts of power and a frequency of 28 kHz for 15 to 60 seconds at sonication temperatures of 20°C and 40°C. In addition, thermoelectric Peltier (12V and 6A) was used to maintaining temperature stability, either as a heater or a cooler.

2.3. Aqueous extraction and separation.
The mixture of bran and solvent was stirred using a magnetic stirrer (500 RPM) for 60 minutes at 60°C. The mixture was then filtered with a mechanical press, and the filtrate was centrifuged for 60 minutes at 4,500 RPM. Following this, the supernatant was stored in the refrigerator overnight and evaporated until the remaining oil and then weighed.

3. Results and Discussion

3.1. Effect of sonication time on yield.
Figure 1 shows the relationship between the RBO yield and sonication time at SLR 0.2 gr/ml.
These results show that sonication time has a positive effect on increasing the RBO yield. There is an enhancement in the sonication period 15 to 60 seconds in yield of 11.8% to 14.7% at 20°C and 16.1% to 17.6% at 40°C. The lifting yield occurs due to the sonication time long enough to maximize the cavitation microbubbles formation, intensifying the solute's mass transfer to the solvent [19]. A related study was conducted by Phan et al. [9] on RBO extraction using the ultrasound-cold press extraction (UCE) at 10 to 45 minutes sonication time and sonication power of 2.25 - 6.75 W/gr. Furthermore, the extraction of RBO from soapstock rice bran with ultrasound-assisted solvent extraction (UASE) was studied by Phan et al. [25] for 4 to 26 minutes and 0.5 to 4.5 W/gr.

Although the sonication time is relatively short in this research, increasing the time may reduce RBO yield. It is based on the results conducted by Khoei and Chekin [18] on RBO extraction using UAAE at 45°C. Huang et al. [17] investigated RBO extraction using ultrasound enzymatic extraction (UEE) and solvent extraction. The RBO yield reduction occurs when sonication takes longer than 60 minutes.

3.2. Effect of solid/liquid ratio (SLR) on yield.

Figure 2 summarizes the RBO yield's relationship with SLR at sonication temperature of 20°C dan 40°C.
At sonication times 15 and 45 seconds (figure 2a), SLR increase can result in reducing the RBO yield from 12.0% to 10.6% (15 seconds sonication time) and 13.4% to 10.8% (45 seconds sonication time). However, at the 60 second extraction time, the RBO yield increase appears at SLR 0.1 - 0.2 gr/ml (13.8 to 14.7%), but declines at SLR 0.2 - 0.3 gr/ml (14.7% to 11.1%). During the short sonication time, the effect of cavitation does not affect SLR. The solvent is not able to compensate for the mass of rice bran and makes a lower yield. Nevertheless, at longer sonication times, the cavitation intensity greatly influences the RBO yield, notably in SLR under optimum conditions. The solvent causes lowering the the RBO yield to not increase the solute's diffusion from the material matrix to the solvent. The higher mass of rice bran influences the mixture's viscosity and resulting resistance in the diffusion process [26].

The pattern at a sonication temperature of 20°C is identical to sonication at 40°C. It concludes that SLR 0.2 gr/ml is the optimum SLR. Similar results were also shown by Djaeni and Listyadevi in RBO extraction using UAE, n-hexane solvent, and extraction temperature of 60°C. They discovered that the optimum bran to the solvent ratio in the extraction was 1: 5 in the range of 1: 2 to 1:15. Similar results were also shown by Mohammadpour et al. [27] on the extraction of Moringa peregrine oil by the UAE and n-hexane solvent at sonication time of 15 minutes, sonication temperature of 40°C, and sonication power of 174 W. According to them, in the liquid-solid ratio range of 5 - 30 ml/gr, oil yields the highest obtained at 20 ml/gr.

### 3.3. Effect of sonication temperature on yield.

Figure 3 shows the correlation between the RBO yield and sonication temperature at 60 seconds of sonication time.
Based on the figure above, the increasing sonication temperature results in the RBO yield on all SLR variables. An increase of the RBO yield at SLR 0.1; 0.2; and 0.3 gr/ml respectively are 3.1%; 19.9%; and 14.2%. Accumulating the sonication temperature can trigger a decrease in the solvent's viscosity and surface tension, reducing the resistance between the solvent and the material. It creates an increasing yield caused by the mass transfer of a solute from the material matrix to the solvent [24]. However, Khoei and Checkin [18], presented different results. Accumulating sonication temperature produces a declining the RBO yield. The same conclusion was presented by Mohammadpour et al. [27].

Huang et al. [17], Xu et al. [16], Senrayan and Venkatachalam [26], Senrayan and Venkatachalam [28] investigated that the yield would increase with increasing extraction temperatures. However, at optimum temperatures, the yield can decrease.

3.4. Effect of sonication treatment on yield.

The relationship between the RBO yield and sonication treatment is presented in figure 4.

Figure 4 shows the effect of sonication treatment on aqueous extraction on the RBO yield. This figure concludes that sonication treatment at high temperature (40°C) increases the RBO yield significantly at 19.79% when compared without sonication treatment as in table 1. Khoei and Chekin [18] also found similar results. In the AE, the RBO yield cannot reach 5%, while in the presence of sonication, it increases more than 20% of the yield. The same was done by Xu et al. [16] investigating RBO extraction using ultrasound-assisted extraction and organic solvents. In research conducted by Phan et al. [9], the sonication effect on RBO extraction with cold press extraction increased yield to 12%. The induction of ultrasonic waves in solvents creates cavitation microbubbles. Due to the mechanism of compression and decompression of microbubbles, it produces high pressure and temperature when encountering a microjet explosion to destroy the cell walls of the material matrix and facilitate mass transfer [19].

Besides, the low temperature (20°C) sonication treatment does not affect the RBO yield. RBO yield tends to relate without using sonication treatment. At low temperatures, there is a significant mass transfer resistance due to the solvent's high viscosity so that it hinders the process of solute diffusion, and the intensity of shear energy, cavitation, and turbulence of the material matrix is low [19].

3.5. Effect of sonication treatment on energy consumption.

Table 1 compares energy consumption in RBO extraction with aqueous extraction method and UAAE at 20°C and 40°C.
Table 1. Energy consumption comparison

| Methods       | Yield Difference | Total Energy Consumption (kWh) | Difference | Relative Energy Consumption (kWh/gr) | Difference |
|---------------|------------------|--------------------------------|------------|------------------------------------|------------|
| AE (reference)| 0                | 1,078                          | 0 %        | 73.31                              | 0 %        |
| UAAE 20°C     | -0.13%           | 1,085                          | 0.65%      | 73.89                              | -0.79%     |
| UAAE 40°C     | 19.79%           | 1,085                          | 0.65%      | 61.61                              | 15.95%     |

The presence of sonication treatment promotes the rise of the RBO yield, as well as energy consumption. Fortunately, progressing the RBO yield is not significant to elevate the required energy. At UAAE 40°C, with an increasing energy utilization of nearly 1.0%, it yields lower relative consumption than AE nor UAAE at 20°C.

4. Conclusion

RBO extraction was successfully carried out using the modified UAAE. The RBO yield was influenced by sonication time and temperature, as well as SLR. In general, the sonication time and temperature contributed positively to significant increase of the RBO yield. However, an increase in SLR did not always promote yield. The highest RBO yield of 17.6% obtained at sonication time of 60 seconds, SLR 0.2 gr/ml, and sonication temperature of 40°C. UAAE is a promising method contrasted to conventional methods, with relatively low energy consumption and produce the highest yield. The development of a combination method, both the conventional method and advance technology on a larger scale is needed to determine the economies of scale of the RBO industry.

Acknowledgement

This research was supported financially by the Kalimantan Institute of Technology through a Participatory Research grant in the 2020 fiscal year (number 2797 / IT10.II / PPM.01 / 2020).

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