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The Effects of Conventional and Microwave Heating Techniques on Extraction Yield of Orthosiphon Stamineus Leaves

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The heating technique in a solid-liquid extraction system plays a significant role in the design and economic potential for the extraction of active components from herbs. This paper focused on the effects of extraction parameters such as ratio of sample to solvent, temperature and time of processing on the extraction yield of Orthosiphon stamineus leaves in conventional and microwave heating extraction techniques. The extracts were concentrated and dried using a rotary evaporator and freeze dryer in order to relate the yield to the processing parameters quantitatively in both heating techniques. The analysis results revealed that the processing parameters; ratio of sample to solvent, temperature and time of extraction had essential effects on the extraction yield of Orthosiphon stamineus leaves. Microwave heating extraction produced a comparable yield to conventional heating extraction with a relatively small deviation of approximately 2.8 % in average. Furthermore, microwave heating extraction reduced processing time, where this technique required about 25 % of the conventional heating time in heating up the extraction mixture to set-point temperature (60 ºC). This study concludes that microwave heating extraction, which is a green technology, has great potential in reducing the carbon foot print due to a shorter processing time and reduced energy consumption (~77 % less) compared to conventional heating extraction.

1. Introduction

Due to increasing health concerns and government support in product development of phytochemicals, the profile of Malaysian herbs has increased significantly over the last 15 years. Since 2000, considerable number of active components in local herbs has been identified and was used in nutraceutical and pharmaceutical products. Malaysian herbs such as Tongkat Ali (Eurycoma longifolia), Kacip Fatimah (Labisia pumila), Hempedu Bumi (Andrographis paniculata), Dukung Anak (Phyllanthus niruri/amarus) and Misai Kucing (Orthosiphon stamineus) had been prove to be among the most useful local herbs due to their physiological benefits or their ability to provide protection against chronic diseases. Orthosiphon stamineus (OS) as example has a number of active components, such as terpenoids, sterols, phenolic acids and lipophilic flavonoids. Furthermore, sinensetin, eupatorin and 3'-hydroxy-5,6,7,4'-tetramethoxyflavone are the main polymethoxylated flavones and rosmarinic acid is the major phenolic acid in OS leaves (Akowuah et al., 2005). The OS leaves tea is traditionally taken as supplement drink for treatment of kidney stone, bladder inflammation, gout, diabetes (Akowuah et al., 2005), edema, influenza and jaundice (Akowuah and Zhari, 2010). In industry, these herbs are typically processed (drying/extraction/leaching) by conventional, indirect heating with an external heat source such as steam or heating oil. For instance, active components in OS such as sinensetin and rosmarinic acid can be commercially extracted using conventional thermal extraction technique such as maceration and Soxhlet extraction. In this extraction process, the surface area of dried OS

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leaves is increased through grinding process and then soaked in a solvent to releases its soluble active components (Cravotto et al., 2011). Thermal extraction technique generally required large volumes of solvent and long processing time due to sluggish heat transfer through conductive/convective mechanisms that depend on thermal diffusivity properties of the heating media and vessel surface for the thermal energy to transfer into the extraction mixture. This extraction technique is relatively slow, inefficient and possibly will cause thermal decomposition of the OS active components compare to non-conventional heating extraction technique such as radio frequency and microwave heating extractions. The main advantage of microwave heating extraction is volumetric heating, where electromagnetic energy is converted to heat directly inside the OS extraction mixture via interactions between the mixture and the electric field. This result in rapid heating and can have significant impacts on reducing processing times (Metaxas and Meredith, 1983). Microwave heating can offer compactness of equipment and selective heating (Ahmad Zaini and Kamaruddin, 2013). In microwave assisted drying for example, energy can be targeted directly at the water molecules within the depth of a solid material and not wasted on the solid itself. This can lead to significant improvements in the energy efficiency of a process (Gabriel et al., 1998). So far as concern, no such study specifically focus on microwave heating extraction of OS leaves has been published except paper related to combination of microwave and ultrasound-assisted in extraction of bioactive from OS (Chung et al., 2017) and dielectric properties for extraction of OS leaves to quantitatively relate the dielectric properties to microwave heating mechanisms and design of microwave applicator (Kamaruddin et al., 2017). The aim of this paper is to perform a preliminary study to investigate and quantify the influence of conventional and microwave heating techniques on extraction of OS leaves. The understanding gained on microwave heating and extraction dependent parameters will provided a basis for the design of a continuous process (potential for scale up). This article was structured with introduction on microwave heating extraction of plant materials, techniques used in conventional and microwave heating and followed by results and discussion and conclusions. In results and discussions, effect of the processing parameters such as extraction time, temperature and ratio of solvent to sample on conventional heating extraction of OS leaves were initially discussed. Later, the discussion focus on comparison study between microwave and conventional heating techniques on extraction yield. This part involved extraction yield, temperature profile and energy consumption against heating time in both heating techniques.

1.1 Microwave heating extraction of plant materials

Various plant materials have been used in microwave heating extraction. Some of the examples are Spruce needles (Picea abies) (Tomaniová et al., 2001), Mesua ferrea L. leaves and Jasminum sambac flowers (Nurdin et al., 2006) and Kiwi seed (Cravotto et al., 2011). Based on the research work had carried out by Tomaniová et al. (2001) on Spruce needles, the extraction efficiency obtained by microwave heating extraction was slightly lower than the conventional thermal heating techniques (Soxhlet extraction). The study was revealed that the utilisation of higher microwave irradiation output power produced higher recoveries of the extracts. However, a longer extraction times did not increase the extraction efficiency in microwave heating extraction. On the other hand, a research done by Nurdin et al. (2006) shows that the extraction time and energy consumption for microwave heating extraction system was eight times lesser than the conventional heating extraction system. Microwave heating extraction system used by the authors was able to produce a higher percentage yield of oil from Jasminum sambac, which is the valuable product. In parallel with results stated by Nurdin et al. (2006), Cravotto et al. (2006) reported that the results of fatty acid composition of the Kiwi seed oil obtained from a microwave heating extraction were similar to a thermal heating extraction (Soxhlet extraction). In terms of extraction time, microwave heating extraction was accelerated the process without inducing obvious changes in the Kiwi seed oil composition. Based on this review, most of microwave heating extraction studies had shown promising results in order to intensify the extraction yield, processing time and energy efficiency in extraction of plant materials. These studies have shown that the microwave heating technique has a great potential to produce a comparable results in extraction of OS leaves.

2. Experimental

2.1 Materials

OS leaves with about 500 (±50) microns in size and distilled water (solvent) were obtained from Center of Lipids Engineering and Applied Research (CLEAR), Universiti Teknologi Malaysia.

2.2 Experiment

The standard extraction procedure used in this study is describes as follow: about 10 g of OS leaves was introduced into a 250 mL conical flask with 200 mL of distilled water for 1 : 20 sample to solvent ratio. This mixture was mechanically stirred until homogeneous, then heated to extraction temperature (i.e. 50 - 90 °C) by use of either an oil bath or a microwave applicator and rigorous stirring was continued throughout the extraction time (i.e. 15 - 150 min). The water based extracts were then carefully filtered using a vacuum pump filter and concentrated using a rotary evaporator. The concentrated extracts were dried in a freeze-dryer until the moisture content less than 5 wt%. The percentage yield of extraction was calculated using Eq(1) as stated below:

$$\text{Extraction yield (wt\%)} = \frac{\text{Mass of dried extract (g)}}{\text{Mass of sample (g)}} \text{ x } 100\ \% \tag{1}$$

2.3 Conventional heating technique

The extraction vessel was immersed in an oil bath that was heated and thermostatically regulated by an IKA hot plate model C-MAG HS7, which controlled the temperature within about ± 1 ºC. The degree of agitation of the extraction mixture was maintained using a magnetic stirrer. Rigorous agitation (~360 rpm) was applied to maintain homogeneity of the mixture and to eliminate temperature gradients inside the mixture. An on-line temperature monitoring system, consist of a data logger (TC-08 Pico Technology) and thermocouple (type K) were used to record the temperature of the mixture through insertion the thermocouple via the neck of the vessel.

2.4 Microwave heating technique

Microwave heating system used in this study utilises a multi-mode cavity excited at 2.45 GHz with a maximum power output of 800 W generated by the magnetron. The microwave system equipped with mode stirrers (rotating blades) to mix up the modes of microwave energy by reflecting waves off the blades and continuously redistribute the electromagnetic field into cavity. Output power calibration based on water load experiment was carried out according to International standard IEC 705 (1988). The absorbed power was determined about 66.9 % (535 W) of the power generated by the magnetron. The power absorbed by the water load is then assumed to be equal to the power absorbed by the extraction sample where the water was the major constituent (> 90 vol%).The system is equipped with an on-line temperature monitoring system, which monitors the bulk temperature conditions of the extraction mixture. The monitoring system consists of grounded copper tube shield thermocouple (type-K), data logger (TC-08 Pico Technology), PicoLog data acquisition software and personal computer. The accuracy in temperature measurements was ± 1.0 °C with 0.1 ºC resolution and calibrated with an infrared thermometer (Fluke-62 Max Plus). The temperature data measured by non-contact infrared sensor on surface of the conical flask is set up in a feedback control loop with the magnetron to regulate the power output to maintain the temperature set point through the on-board processor.

3. Results and discussions

3.1 Effect of the processing parameters on conventional heating extraction

In order to assess the repeatability and evaluate the time dependence of the extraction of OS leaves conducted using the conventional heating technique, three duplicate experiments under the same conditions (sample: solvent = 1 : 20) for each specific extraction time (i.e. 30, 60, 90, 120 and 150 min) were carried out a constant temperature, 60 ºC (± 1 °C). Figure 1a shows the plots of yield against extraction time for the repeatability and time dependence assessment experiments discussed above. The experimental results are expressed together with the error bars which is produced an estimation error about 8.4 % in maximum. Based on these results, it was concluded that the conventional heating extraction carried out in this research study are repeatable and reliable. For the time dependence assessment, the lowest yield was obtained at 60 min at 17.9 %, while the highest yield was at 30 min at 22.9 %. Based on this result, the best time of extraction is about 30 min meanwhile prolonging the extraction time gave no significant increase on extraction yield.

To evaluate the temperature dependence of the extraction progress of OS leaves in the conventional heating technique, five extraction mixtures with identical sample to solvent ratio (1 : 20) and masses were prepared via the procedure mentioned in Section 2.2 and these mixtures were extracted at 50, 60, 70, 80 and 90 ºC for 90 min. The temperature studied was limit to 90 °C to avoid solvent (distilled water) loss due to evaporation. Figure 1b shows the yield plotted against extraction temperatures. It can be seen from Figure 1b that the lowest yield was obtained at 90 ºC at 17.7 %, while the highest yield was at 70 ºC at 22.2 %. As the temperature of extraction increases, the yield also increases due to the higher energy supplied which makes the solute and solvent more energetic to mix and diffusion process between both materials become quicker. Although, this is true from 50 ºC to 70 ºC, but above that, the yield started to decline. This is believed due to some amounts of the solvent were lost to the surrounding through evaporation for the extraction carried out above 70 ºC.

The effect ratio of sample to solvent on extraction of OS leaves using conventional heating method was investigated at a fixed temperature and time. The extraction temperature was fixed at 60 °C (± 1 °C) to minimise solvent loss due to evaporation for 90 min extraction time. The sample to solvent ratio was examined at 5 different ratios, which is 1 : 7, 1 : 10, 1 : 20, 1 : 30 and 1 : 40. Figure 1c shows the yield of extract versus ratios of sample to solvent. The highest yield was obtained at ratio 1 : 20 with 14.5 %, while the lowest was at ratio 1 : 7 with 7.4 %. The sample with ratio 1 : 7 is also faced difficulty in handling during the filtration process due to easily clog the vacuum filter. The yield for ratios 1 : 20 and 1 : 30 are 14.5 % and 14.0 %. The difference in yield between the two mixtures (1 : 30 and 1 : 40) to the 1 : 20 mixture is quite small and it can be said that the mixture is nearly saturated at these points.

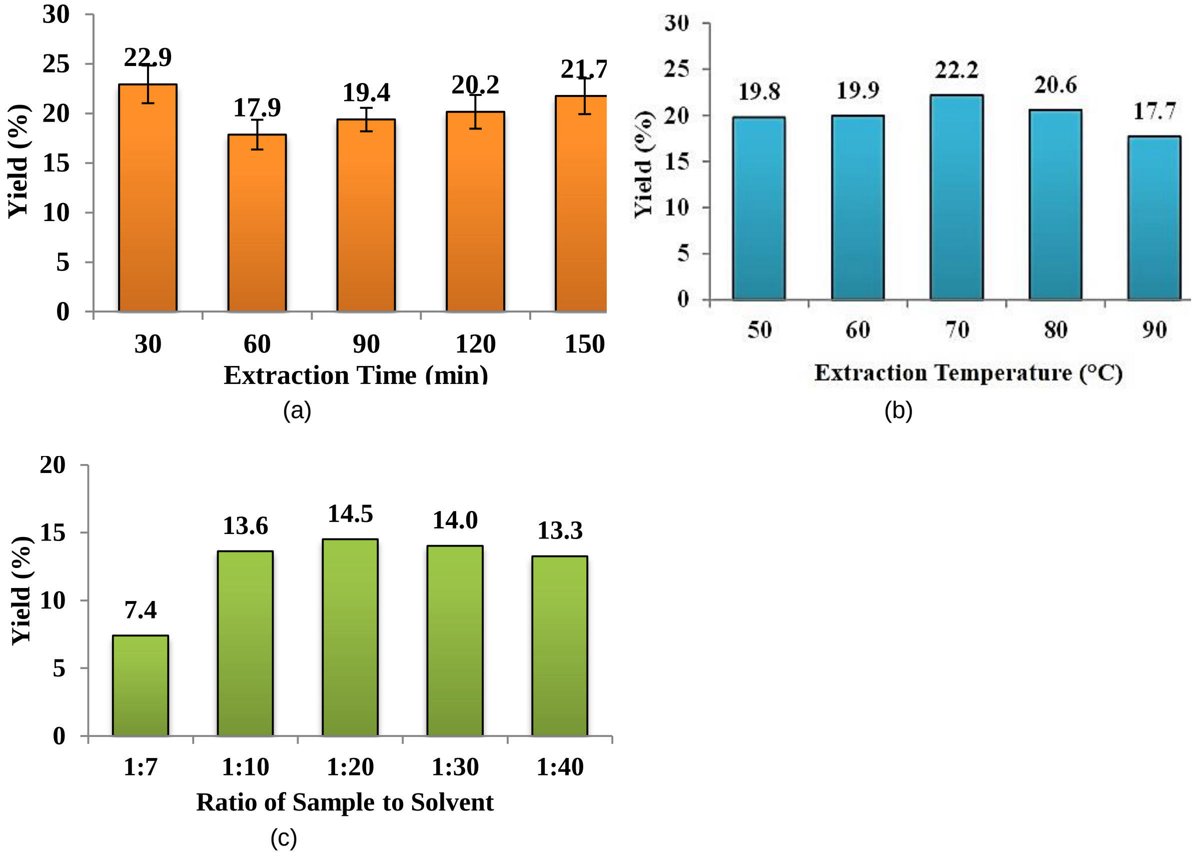

Figure 1: Extraction yield against processing parameters: (a) extraction time, (b) extraction temperature, and (c) ratio of sample to solvent. The maximum estimated standard deviations of the measured yields are ± 2 %

3.2 Comparison study between microwave and conventional heating techniques on extraction yield

In order to compare the yield behaviour of the extraction of OS leaves using conventional thermal and microwave heating, a study utilising both heating techniques was carried out and directly compared. The extraction system for fixed sample to solvent ratio (1:20) at 60 °C changes with extraction time (15 - 90 min) was studied. The plots of yield versus the extraction time of both heating techniques are shown in Figure 2a.

Based on results obtained in Figure 2a, microwave extraction produced comparable yields against conventional extraction at each experiment. The highest yield for microwave extraction was at 90 min with 19.5 %, while the lowest yield was at 15 min with 8.9 %. The highest yield for conventional extraction was at 30 min with 22.9 %, while the lowest yield was at 15 min with 11.6 %. The average difference of the yield between the two methods is about 2.8 %, which is relatively small and reliable. On top of that, it can also be observed that prolonging the extraction time in both heating techniques produced nearly plateau yield of extraction. Thus, it can be said that the ideal time of extraction for both heating techniques is about 30 min.

The temperature profiles against heating time for both techniques on 15 min extraction study are shown in Figure 2b. The time taken for the conventional heating to achieve the set-point temperature 60 °C was about 800 s (~13 min). Meanwhile, the microwave heating technique with average output power 500 W only took less than 200 s (~3 min) for a similar target temperature. The former technique takes longer time because the heat from a hot plate (oil bath) is slowly transferred to the extraction mixture through conduction and convection mechanisms. In contrast with the latter, microwave energy is directly transfer into the mixture at nearly speed of light and heat is generate in-situ through a unique heating mechanism that called dipoles rotation (Hayes, 2002). Based on heating mechanisms explained, conventional heating extraction produced a little bit higher yields because the overall contact time between solvent and OS sample is longer compared to microwave heating extraction, where the experimental time of extraction is only considered when the set-point extraction temperature is reached. Microwave heating extraction also indirectly reduces the processing time.

Figure 2c shows the energy consumption for conventional and microwave heating extractions determined based on the input power recorded by a power meter and microwave power absorption (sample load) experiment. The figure indicates that microwave heating extraction consumes less energy about 13 % and 52 % from the conventional heating extraction in heating up process (ambient to set-point temperature) and maintain the extraction temperature (60 ºC). This result is in good agreement with previous studies in extraction of of essential oils (Nurdin et al., 2006), natural products (Desai et al., 2010), and herb, Eurycoma Longifolia (Foong,et al., 2015) which stated that the microwave heating extraction is energy efficient compared to the conventional heating extraction.

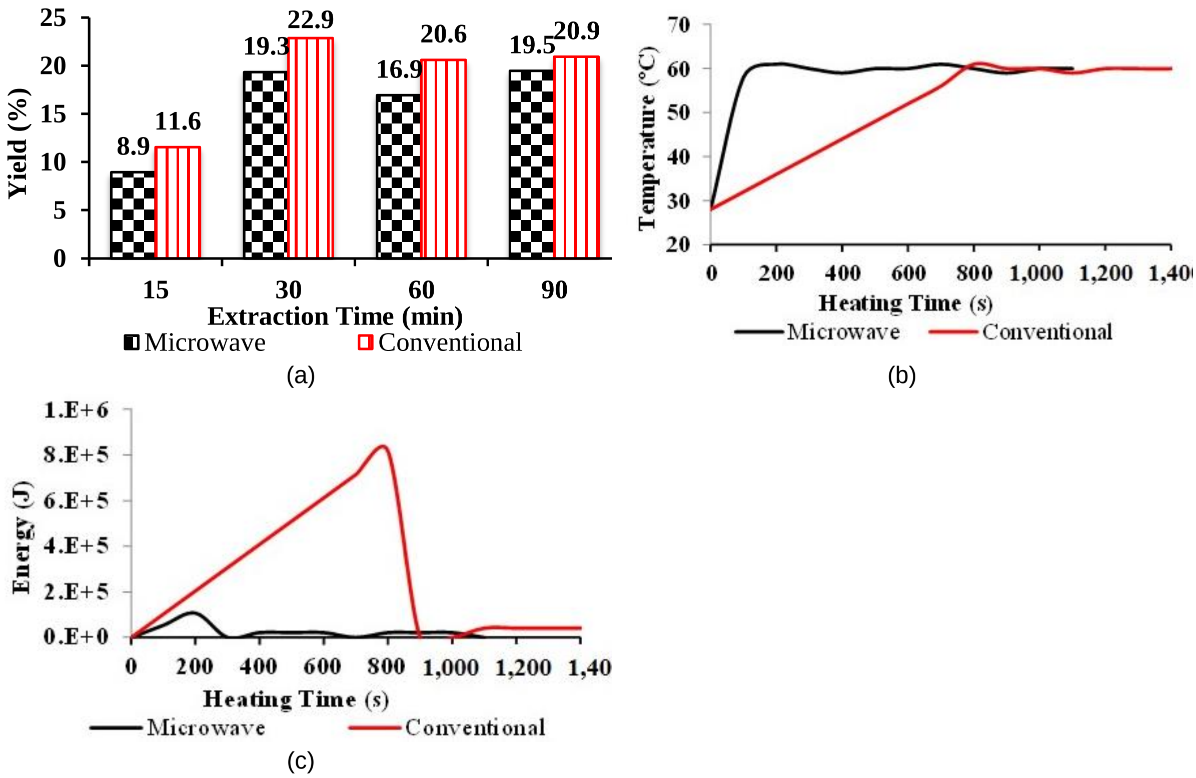

Figure 2: Comparison study for microwave and conventional heating techniques (a) yield vs. extraction time, (b) temperature vs. heating time for 15 min extraction, and (c) energy vs. heating time for 15 min extraction. The maximum estimated standard deviations of the measured data: Yield ± 2 %, Temperature ± 1 ºC.

4. Conclusions

The processing parameters which affect the extraction yield of OS leaves using conventional thermal heating technique were the time and temperature of extraction and mass ratio of OS sample to the solvent used. For an assessment study of heating techniques on OS leaves extraction, the microwave heating produced reliable results and comparable yields to the conventional thermal heating. Microwave heating extraction has a great potential as an alternative to the conventional thermal extraction method in the aspects of time and energy consumptions. The microwave heating technique provided a significant saving (~77 %) in heating-up time and energy consumption of the OS leaves extraction.

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