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ULTRASONIC-ASSISTED EXTRACTION OF OIL FROM *CALOPHYLLUM INOPHYLLUM* SEEDS: OPTIMIZATION OF PROCESS PARAMETERS

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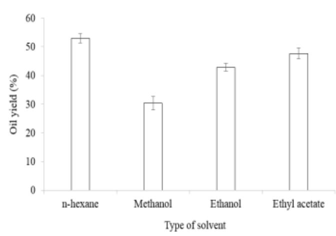
Graphical abstract



Abstract

Calophyllum inophyllum (*C. inophyllum*) is a plant known as "Penaga Laut" in Malaysia. Its seeds contain valuable oil that serve as a potential alternative sources for vegetable oil. In this study, *C. inophyllum* seeds oil was extracted using an ultrasonic-assisted extraction (UAE) technique. The optimization of extraction parameters namely different types of solvent, extraction time, ultrasonic power, extraction temperature and liquid to solid (L/S) ratio were performed using one-factor-at-a-time (OFAT) method. The optimum extraction conditions obtained were *n*-hexane as a solvent, extraction time 20 min, ultrasonic power 210 W, extraction temperature 40°C and L/S ratio 20 ml/g, with an oil yield 55.44 ± 0.53 %.

Keywords: *Calophyllum inophyllum*, seed oil, ultrasonic-assisted extraction, optimization

Abstrak

Calophyllum inophyllum (*C. inophyllum*) ialah sejenis pokok yang dikenali sebagai "Penaga Laut" di Malaysia. Ia mengandungi minyak yang berpotensi sebagai sumber alternatif bagi minyak sayuran. Dalam kajian ini, minyak daripada benih *C. inophyllum* telah diekstrak dengan menggunakan teknik pengekstrakan ultrasonik. Pengoptimuman bagi proses parameter iaitu jenis pelarut yang berbeza, masa pengekstrakan, kuasa ultrasonik, suhu pengekstrakan dan nisbah cecair kepada pepejal (L/S) telah dijalankan dengan menggunakan kaedah satu-faktor-pada-satu-masa (OFAT). Keadaan pengekstrakan optimum yang didapati adalah *n*-hexana sebagai pelarut, masa pengekstrakan 20 minit, kuasa ultrasonik 210 W, suhu pengekstrakan 40°C dan nisbah cecair kepada pepejal 20 ml/g, dengan hasil minyak 55.44 ± 0.53 %.

Kata kunci: *Calophyllum inophyllum*, seed oil, ultrasonic-assisted extraction, optimization

1.0 INTRODUCTION

Calophyllum inophyllum Linn. (*C. inophyllum*) is an oilseed ornamental evergreen tree, belongs to the Clusiaceae (Guttifereae) family [1]. It is commonly known as "Penaga Laut" in Malaysia and "Alexandrian laurel" in English [2]. The genus *Calophyllum* comprises of about 180-200 species distributed in many tropical areas around the world [3]. *C. inophyllum* is a beautiful plant native to India, East Africa, South East Asia, Australia and South Pacific [4]. The plant is widespread from the Indian Ocean such as Africa and India; throughout Malaysia and in the Pacific islands [5]. It is a medium sized tree that grew up to 20 m high having a broad spreading crown of irregular branches [6, 7]. The light grey bark shows deep fissures alternating with flat ridges [4]. The leaves have opposite arrangements, and are petiolate, thick and shiny with numerous parallel secondary veins while the flowers are borne in axillary cymes, white, and fragrant, with variable numbers of perianth parts and yellow anthers [5]. The round-shaped, light green fruits which are wrinkled upon ripening contain a single seed with very high oil content (75 %) [8]. The oil which also known as domba oil, pinnai oil or dilo oil is greenish yellow in colour with an aromatic odour and insipid taste [9]. It is used for treating various skin diseases including scabies, ringworm and dermatosis [1]. This oil is also found to exhibit anti-human immunodeficiency virus (HIV), antimicrobial and cytotoxicity properties [10, 11]. In recent years, *C. inophyllum* oil have been recognized as a potential source for biodiesel [12].

Since ancient times, plant materials such as juices, gums, fatty oils and essential oils are considered as a rich source of natural, effective and safe medicines [13]. Fatty oils obtained from vegetable sources are normally isolated from the seeds of oleaginous plants, cereals and fruits [14, 15]. Nut oil, seed oil and oil of fruits and vegetables are receiving significant attention due to their high concentration of bioactive lipid components, such as polyunsaturated fatty acids and phytosterols, which have shown various health benefits [16]. They have been considered as alternatives to treat infectious diseases and some of them have documented to possess pharmacologically important activities like antimicrobial, antifungal and antitumor properties [17, 18]. In addition, seed oils are also beneficial in the prevention and treatment of several diseases including arthritis, diabetes and hypertension [19]. These oils do not only provide skin protection against reactive oxygen species, but also presented significant antipyretic, analgesic, anti-carcinogenic and anti-inflammatory activities [20, 21]. Vegetable oils represent promising alternatives to conventional fuel because of their similar properties [22]. Such oils include soybean oil, palm oil, sunflower oil, safflower oil, rapeseed oil, coconut oil and peanut oil [23]. The use of natural oils as a biodiesel offers many advantages because they are non-toxic, renewable, biodegradable and has a high flash point [24, 25]. In the production of polymers, vegetable oils have been identified as potential substitutes for the petrochemical derivatives [26]. They find innumerable industrial applications such as plasticizers, lubricants, adhesives, biodegradable packaging materials, printing inks, paints and coatings [27]. Moreover, plant oils also play an important role as food preservatives in the food industry [28].

The use of plant oils have become increasingly important for scientific research and industrial applications [29]. Plant oils are one of the important products of agriculture based industry such as biodiesel, food, pharmaceutical and cosmetic industries which have become a multibillion dollar international market [30-32]. As a consequence, the global and national markets for natural oils have been growing rapidly and significant economic gains are being realized [33]. Therefore, more alternative sources of vegetable oil from underutilized plant crops need to be explored in order to fulfill the increasing demands of vegetable oil.

Most of the oil originated from oil-producing plants especially seeds, were extracted conventionally by using a mechanical press or organic solvent extraction [34]. The oil obtained by mechanical process is of high-quality, however, the process consumed high energy, required high level equipment and produced lower extraction yield [35, 36]. In fact, it is a time and labour intensive process [7]. Even though this traditional technique was relatively inexpensive, it was not as efficient as compared to solvent extraction [37]. Chemical extraction which used organic solvents such as *n*-hexane, light petroleum ether, and a mixture of chloroform-methanol has become an alternative technique in isolating oil from plant seeds with a simpler process and higher oil yield [38]. Unfortunately, the usage and disposal of great amount of organic solvents which leads to environmental problems and economical inconveniences have caused the organic solvent extraction method to be gradually abandoned [36, 39]. Supercritical fluid extraction (SFE) technology represents an efficient alternative which gives a higher extraction yield than that of traditional methods. The CO_2 solvent used in SFE is known for its non-polluting, non-toxic and inexpensive properties, making SFE an environmental-friendly technique [40]. However, the applicability of this technology at a larger scale is limited due to its relatively high investment, operating and maintenance costs [41, 42]. Microwave-assisted extraction (MAE) offers higher extraction yield than the solid–liquid extraction technique [41]. However, the efficiency of MAE can be poor when the target compounds or solvents are non-polar, or when they are volatile; and the use of high temperatures that can lead to degradation of heat-sensitive bioactive compounds [43]. In such a case, it would be beneficial to establish an improved technique of extracting oil from plants that are equally competent and at the same time economically viable.

Ultrasonic-assisted extraction (UAE) has emerged as a potential approach to extract valuable substances from plant raw materials [44]. It has been found to be useful in increasing the yield and mass transfer in many solid – liquid extraction processes [45]. UAE can be applied in two different techniques, either by using an ultrasonic bath or an ultrasonic horn transducer [46]. Ultrasonic bath which was equipped with a temperature-controlled device facilitated the recovery of thermo-sensitive compounds including essential oils [47]. The improvement of extraction process caused by ultrasound was attributed to the propagation of ultrasound pressure waves which resulted in the acoustic cavitation phenomenon [48]. This phenomenon was responsible for the disruption of cell walls, reduction of particle size and increasing the contact surface area between solid and liquid phases [49]. In fact, ultrasounds also exerted a mechanical effect which promotes better penetration of solvent into the sample matrix thus allowing higher diffusion rates across the cell wall [50, 51]. According to Jovanovic-Malinovska *et. al.* [52], UAE allows higher extraction yield, higher rate of extraction, shorter extraction time and higher processing throughput along with the benefits of less solvent consumption and lower extraction temperature which was advantageous for the extraction of heat labile compounds. Recently, UAE was used widely in the extraction of oil from various type of seeds including tobacco [53], papaya [54] and black seeds [55]. This technique has also been used in the extraction of bioactive components from natural products such as phenolic compounds, essential oils, polysaccharides and others [56].

Few researchers have reported on the extraction of *C. inophyllum* seed oil using various techniques. Hathurusingha *et. al.* [57] used solvent extraction with *n*-hexane to extract oil from *C. inophyllum* seeds collected from three different northern Queensland provenances (Cardwell, Townsville, Yeppoon) and reported that the oil content was in the range of 29.86 % to 46.17 %. In another study, *C. inophyllum* seed oil were extracted using both screw pressing and solvent extraction techniques [7]. The highest oil yield obtained from the former and latter technique were approximately 25 % and 51 %, respectively. In this study, UAE was applied as an alternative approach in extracting oil from *C. inophyllum* seeds as well as to attain the optimum yield of the oil.

It is necessary to study the influence of every process variables towards the response variable in order to achieve an optimum oil extraction conditions which is useful for scale up and industrial processing purposes [58]. Therefore, the objective of this study is to optimize the UAE conditions of *C. inophyllum* seed oil using OFAT method. Five extraction process parameters considered were different types of solvent, extraction time, extraction temperature, ultrasonic power and L/S ratio.

2.0 EXPERIMENTAL

2.1 Preparation of Raw Materials

Matured *C. inophyllum* fruits were collected from the ground under the tree located in Taman Kerian, Parit Buntar, Perak, Malaysia. The identification of the species was provided by the School of Biological Sciences, Universiti Sains Malaysia (USM. Herbarium 11565). The fruits were cleaned and stored at ambient temperature. Prior to oil extraction, the fruits were manually cracked to obtain the seeds. The seeds were selected according to their condition, only the good-conditioned seeds were used while the damaged seeds were discarded. Then, the cleaned seeds were ground using a dry mill and sieved through a 10-mesh sieve (pore size 2 mm) shaker. The seed samples with particle size of 2 mm and below were used in this study. Chemicals including *n*-hexane (Merck), ethanol (Merck), methanol (Merck) and ethyl acetate (Merck) were used as solvents. All chemicals are of analytical grades.

2.2 Determination of Moisture Content of *C. inophyllum* Seeds

The moisture content of *C. inophyllum* seeds was determined by the oven drying method as described by Bamgboye and Adebayo [59]. 30 g of the cleaned seeds was weighed and dried in an oven at 105 ± 1°C for 24 hr and the weight was recorded after every 2 hr. The procedure was repeated until a constant weight was obtained. After each 2 hr, the sample was removed from the oven and placed in the desiccator for 30 min to cool. It was then re-weighed. The moisture content (wet basis) was calculated based on Equation 1:

$$Moisture\ content\ (\%) = \left(\frac{m_i - m_d}{m_i}\right) \times 100 \qquad (1)$$

where m_i and m_d is the initial and final mass of the seed (g), respectively [60].

2.3 Ultrasonic-Assisted Extraction

Extraction process was performed according to the method as described by a few researchers [55, 61], with slight modification. An ultrasonic water bath (Transsonic Digital S Model T840DH) was used for indirect ultrasonication. It has an internal tank dimension of 327 mm × 300 mm × 200 mm, volume of 18 L, power consumption of 1100 W and a fixed operating frequency of 40 kHz. The ultrasonic bath was equipped with a timer, heater, temperature regulator and indicator, as well as ultrasonic power regulator (70-250 W). The bath was filled with distilled water approximately 2/3 of its volume (about 12 L). During the extraction, the temperature was controlled and maintained at the desired level by circulating distilled water through the ultrasonic bath using a thermo-circulator water bath. The seeds (5 g) and the extracting solvent, *n*-hexane (100 ml) were mixed in an Erlenmayer flask (250 ml) and covered with aluminium foil. The flask was immediately immersed in the ultrasonic bath and fixed at the center position for each experimental run. The extraction was carried out for 15 min at ultrasonic power of 190 W while the bath temperature was kept constant at 25 ºC. After the extraction process was completed, the mixture was centrifuged at 4,000 rpm for 20 min to separate the liquid extract from the solid residue. Subsequently, the solvent was removed from the liquid extract using a rotary evaporator. The extracted oil obtained were dried in a desiccator and weighed until a constant weight was reached. The extraction was repeated using different types of solvent (ethyl acetate, ethanol and methanol), different extraction time (5 min,10 min, 15 min, 20 min, 25 min, 30 min and 35 min), different ultrasonic power (70 W, 110 W, 150 W, 190 W, 210 W, 230 W and 250 W), different extraction temperature (25ºC, 30ºC, 35ºC, 40ºC, 45ºC, 50ºC, 55ºC and 60ºC) and different L/S ratio (5 ml/g,10 ml/g, 15 ml/g, 20 ml/g, 25 ml/g, 30 ml/g and 35 ml/g).

2.4 Determination of C. inophyllum Seed Oil Yield

Extraction yield of *C. inophyllum* seed oil was calculated using Equation 2 [62]:

$$Oil\ yield\ (\%) = \frac{Mass\ of\ oil\ (g)}{Mass\ of\ seed\ sample\ (g)} \times 100 \qquad (2)$$

All experiments were performed in triplicates and the yields obtained represented an average measurement.

3.0 RESULTS AND DISCUSSION

3.1 Effect of Different Types of Solvent

The *C. inophyllum* seeds used in this study have a moisture content of 9.45%. The extraction of oil from *C. inophyllum* seeds was performed using different solvents including *n*-hexane, methanol, ethanol and ethyl acetate. As can be seen in Figure 1, the oil yield depends on the type of solvent used in the extraction process. The highest oil yield of 52.93 ± 1.52 was obtained using *n*-hexane followed by ethyl acetate, ethanol and methanol. According to Rodríguez-Solana et. al. [63], the polarity index of *n*-hexane, ethyl acetate, ethanol and methanol was 0, 4.3, 5.2 and 6.6, respectively. This means that the oil yield decreased as the relative polarity of the solvent increased. The oil from *C. inophyllum* seeds was classified as a non-polar lipid due to their major constituents of oleic acid [64-66]. Therefore, they dissolved better in *n*-hexane.

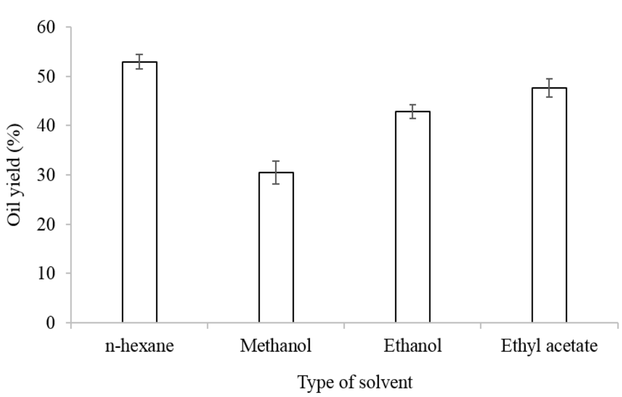


Figure 1 Effect of different types of solvent on C. *inophyllum* seed oil yield. (Conditions: Extraction time 15 min, ultrasonic power 210 W, extraction temperature 40 ºC, L/S ratio 20 ml/g)

3.2 Effect of Extraction Time

Extraction time was another process factor that would influence the extraction yield of *C. inophyllum* seed oil. Figure 2 shows that the oil yield increased exponentially with extraction time until it has reached equilibrium. The oil yield increased from 48.09 ± 0.29 % to 55.44 ± 0.53 % when the extraction time ranged between 5 min and 20 min. This observation could be due to the complete cracking of the seed cells due to acoustic cavitation effects during the early stage of extraction, which lead to a better solvent penetration into the cells and facilitating the release of oil within the cells into the exterior solvent [67, 68]. Zou et. al. [69] stated that the increment of oil yield with respect to extraction time was also due to the increase in number of cavitation micro-bubbles formed by ultrasound.

However, though the extraction was extended until 35 min, the oil yield did not show any further significant improvement, indicating that the oil yield started to maintain a dynamic equilibrium with increasing extraction time. This is in agreement with the results obtained by Dong et. al. [70] who stated that dissolved target oil components tend to re-adsorb on the ruptured seed particles owing to the large specific surface area, thus lowering the yields of oil. Furthermore, when the diffusion front moved towards the interior of the cell tissues, the diffusion area reduced, diffusion distance increased and the diffusion rate would decreased accordingly [67]. Hence, there was no significant changes observed in the oil yield during the prolonged time period. The most suitable time duration for extracting *C. inophyllum* seed oil is 20 min.

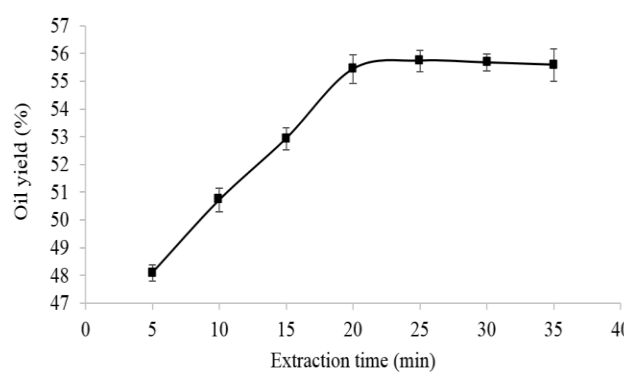


Figure 2 Effect of different extraction time on *C. inophyllum* seed oil yield. (Conditions: *n*-hexane as a solvent, ultrasonic power 210 W, extraction temperature 40 ºC, L/S ratio 20 ml/g)

3.3 Effect of Ultrasonic Power

Figure 3 shows a positive correlation between ultrasonic power rise and the *C. inophyllum* oil yield. The maximum oil yield of 55.44 ± 0.53 % was obtained at 210 W ultrasonic power under the employed operating conditions. The amount of oil extracted increased significantly as the ultrasonic power was raised from 70 W to 210 W. However, a reverse trend was observed when the ultrasonic power was increased above 210 W. A slight decrease in the oil yield was recorded between 210 W and 250 W.

The increment of oil yield was attributed to the formation of cavitation bubbles and violent explosion of those bubbles onto the cell surface [71]. Such phenomenon resulted from a large amplitude of ultrasonic waves travelling through the solvent medium, thus generating a micro-jet which disrupted the cell wall [48, 72, 73]. The cell wall disruption accelerated the solvent penetration into the cells and released of oil into the solvent, hence improving the mass transfer rate [74]. In addition, the vibrating effect from ultrasonic waves might help the oil and the extracting solvent to mix better, thereby increasing the oil extraction efficiency [72].

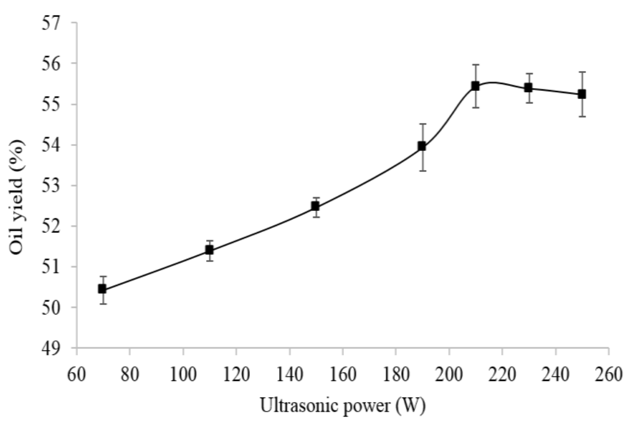


Figure 3 Effect of different ultrasonic power on *C. inophyllum* seed oil yield. (Conditions: *n*-hexane as a solvent, extraction time 20 min, extraction temperature 40 °C, L/S solid ratio 20 ml/g)

On the other hand, the reduction in the oil yield during the greater application of ultrasonic power was probably due to the elevation of the solvent temperature as well as vapor pressure which lead to a drop in the cavitation intensity [62]. This occurrence did not create a conducive condition for the enhancement of the extraction process and improvement of the oil yield. Therefore, 210 W was found to be the most suitable ultrasonic power for the UAE of oil from *C. inophyllum* seeds.

3.4 Effect of Extraction Temperature

The effect of extraction temperature on *C. inophyllum* seed oil yield was shown in Figure 4. It was observed that the oil yield increased with the increase in temperature and reached a peak value at 40°C. When the temperature was increased until 60°C, the oil yield experienced a notable reduction. Higher extraction temperature resulted in a higher solubility of the oil in the extracting solvent and a lower solvent viscosity [75, 76]. This has contributed to an improvement in the mass transfer of the oil into the solvent as well as the solvent diffusion rate into the pores of the seeds [77]. According to Feng et. al. [78], heating effects have weaken the cell wall integrity of the seeds and thereby providing larger contact surface area between the solvent and the oil. As a result, more oil would be extracted into the solvent.

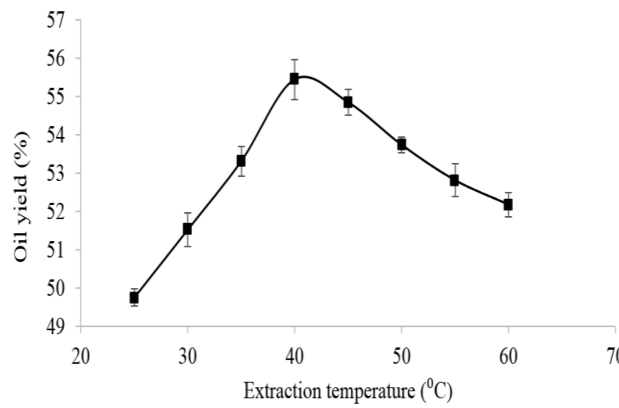


Figure 4 Effect of different extraction temperature on *C. inophyllum* seed oil yield. (Conditions: *n*-hexane as a solvent, extraction time 20 min, ultrasonic power 210 W, L/S ratio 20 ml/g)

Meanwhile, the oil yield did not show further increment when the temperature was beyond 40 °C. This could be due to the formation of vapor-filled bubbles at elevated temperature and as a result, the implosion of cavitation bubbles would be cushioned [79]. This cushioning effect would eventually cause the ultrasound cavitation effect to be less efficient. According to Xu et. al. [80], when the vapor pressure of the solvent was high, the cavitation was achieved at a lower ultrasound intensity, thus reducing the strength of shear forces in the vicinity of the bubbles. Hence, a decrease in the oil yield at higher extraction temperature was expected. The preferable extraction temperature for extracting *C. inophyllum* seed oil is 40 °C.

3.5 Effect of Liquid to Solid Ratio

In this study, the effect of L/S ratio on *C. inophyllum* oil yield was evaluated from 5 ml/g to 35 ml/g. The results revealed that the oil yield increased with increasing L/S ratio and the highest value was obtained at a ratio of 20 ml/g (Figure 5). An insignificant changes in the oil yield was observed when the L/S ratio was increased above 20 ml/g. According to Zhang et. al. [81], the significant increase in the oil yield at a higher L/S ratio resulted from a greater driving force during the mass transfer of oil which caused by a larger concentration gradient between the interior cells and the exterior solvent. This driving force favors diffusion thus allowing the oil to dissolve in the solvent at a higher rate [82].

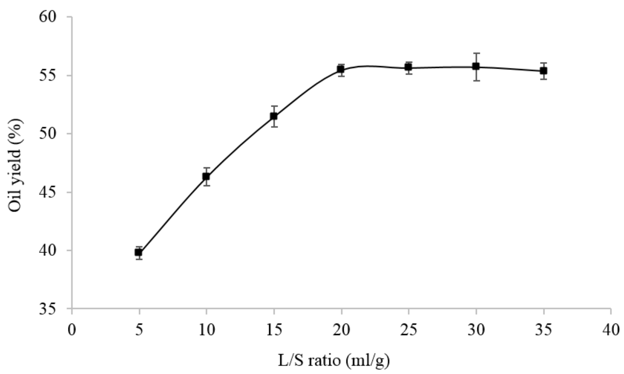


Figure 5 Effect of different L/S ratio on *C. inophyllum* seed oil yield. (Conditions: *n*-hexane as a solvent, extraction time 20 min, ultrasonic power 210 W, extraction temperature 40 °C)

However, beyond L/S ratio of 20 ml/g, the oil yield remained almost unchanged. This can be attributed to the prolonged distance of diffusion towards the interior cell tissues which results in a lower solvent diffusion rate [83]. Moreover, since the limitation to mass transfer was more confined to the solid interior, an excessive amount of solvent would not change the driving force [79]. This means that further increase of L/S ratio would not increase the oil yield. Hence, the most appropriate L/S ratio in the UAE of *C. inophyllum* seed oil is 20 ml/g.

4.0 CONCLUSION

This study reported on the application of ultrasound for the extraction of oil from *C. inophyllum* seeds, which represent a promising alternative source of vegetable oil. The highest oil yield was obtained using *n*-hexane as a solvent. The efficient extraction period for achieving maximum yield was about 20 min. Extraction yield increased with an increase in extraction temperature, ultrasonic power and L/S ratio. Their optimum values were found to be 40°C, 210 W and 20 ml/g, respectively. Under these extraction conditions, the *C. inophyllum* seed oil yield recorded was 55.44 ± 0.53 %.

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