Ultrasonic Assisted Extraction (UAE) of *Moringa oleifera* Seed Oil

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Abstract

This study aims to investigate *Moringa oleifera* seed oil extraction using indirect ultrasonication or known as ultrasonic assisted extraction (UAE). The process was conducted at power of 176kW and ultrasonic frequency of 40 kHz. Usage of highly toxic and carcinogen solvent was avoided in this study by using ethyl acetate as replacement. The influence of extraction temperature, liquid to solid (L/S) ratio and time were looked at. Experimental data reveals that the maximum *Moringa oleifera* seed oil extraction (38.1%) was obtained at conditions; 30°C, L/S 1:20 after 40 mins of extraction. The study was followed by fitting the experimental data using three kinetics empirical models; Power law model, Parabolic diffusion model and Naik model to identify the best fit data. It was found that, Power law model fitted well to the extraction data with R^2 value of 0.9919. Thus, it is adequate to describe the extraction process of oil from *Moringa oleifera* seeds.

Keywords; *Moringa oleifera* ; seed oil ; ultrasonic assisted extraction (UAE) ; model fittings ; optimization ; ethyl acetate

1. Introduction

Moringa oleifera belongs to the family of *Moringaceae*. It is one of the most popular plants in the South East Asia region where the species is native to north western India, and widely cultivated in tropical and subtropical areas (Premi & Sharma, 2013). The plant is usually known as 'drumstick tree' or 'horseradish tree' (Zheng et al., 2017). It grows fast with great economic value and considered very useful, as every part of it has its own uses. Various parts of the plant can be utilized as a source of food, as well as in medicinal application (Mani et al., 2008). *Moringa oleifera* can be cultivated for its leaves, pods or mainly its seed or kernel for oil extraction. The yields vary widely, depending on the season, fertilization and irrigation regimen (Gopalakrishnan et al., 2016). Yielding *Moringa oleifera* seed with different growing condition such as warm, dry and some supplemental fertilizer and irrigation are crucial to promote the best farming practices (Leone et al., 2016). Fig. 1 shows the *Moringa oleifera* pods, and seeds.

The seeds of *Moringa oleifera* are rich source of oil and protein. It contains oleic acid also known as ben oil, antibiotic called pterygospermin, and fatty acids for instance, linoleic acid and behenic acid as well as phytochemicals such as tannins, saponin, phenolics, phytate, flavonoids, terpenoids and lectins. Furthermore, *Moringa oleifera* seeds are rich in fats, fiber, proteins, minerals, vitamins A, B, C and also amino acids which are beneficial to human skin. *Moringa oleifera* seed oil can be used for rheumatism and gout treatment, preparation of cosmetics, lubricant in watch making and precision equipment, purification of blood and enhancing cardiac function as medicine, and also for edible purpose (Gopalakrishnan et al., 2016).

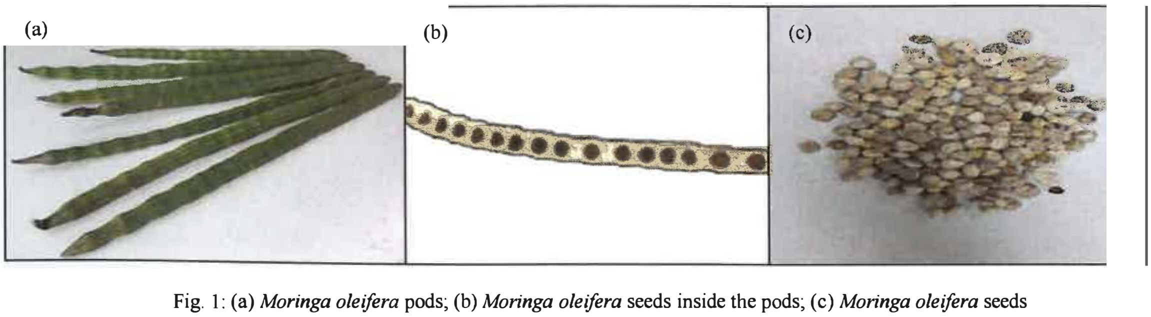

Fig. 1: (a) *Moringa oleifera* pods; (b) *Moringa oleifera* seeds inside the pods; (c) *Moringa oleifera* seeds

To date, there are several established techniques to extract oil from *Moringa oleifera* seeds. They are cold pressing, Soxhlet extraction and ultrasound assisted extraction (UAE). Cold pressing is suitable for extraction under mild process conditions according to Soto et al. (2007) and this process was applied to extract many other types of seed oil such as *Nigella sativa.* (Mohammed et al., 2016), Chia seeds (Ixtaina et al., 2011) and ***Calophyllum inophyllum*** (Kedzia et al., 2011). This method is highly preferred due to the lower extraction temperature requirement which avoid oil oxidation in order to maintain the quality of oil (Siger et al., 2017). However, this extraction method thus avoiding the side extraction yield. In addition to that, the degradation of the active compounds due to uncontrolled of light exposure and oxygen is highly presence and have the lowest oxidative stability (Wroniak et al., 2008).

Soxhlet extraction offers continuous process for solid-liquid extraction. The extractor consists of a side called thimble, where it contains the material to be extracted. This method uses a large amount of organic solvent that is heated in reflux. Unfortunately, Soxhlet extraction is capable for only single extraction and it requires a very long extraction time, typically 8 hours and the extraction yield is reported to be influenced by the condition of the thimble (Halfadji & Touabet, 2013). The extraction usually occurs at the boiling point of the solvent for a long time. So, the possibility of thermal decomposition of the natural substances cannot be ignored.

Ultrasonic assisted extraction (UAE) is an alternative method for oil extraction that allows high extraction yield, shorter extraction time and low consumption of solvent (Hadi et al., 2015). Ultrasonic water bath was used for indirect ultrasonification at typically 176kW and 30% kHz of power and frequency, respectively (de Mello et al., 2017). This method is easy to handle and requires shorter extraction time as well lower consumption of solvent (Halfadji & Touabet, 2013).

However, the study on the application UAE of *Moringa oleifera* seed oil is rarely updated, hence, the kinetics of the oil extraction. The empirical kinetic model has been applied widely in the extraction of natural substances (Veličković et al., 2006). These models are Power law model, Parabolic diffusion model and Naik model which were described by the mathematical variations in the next section. The mathematical models were applied to the extraction of natural product with time. The Power law model mechanism is diffusion of any natural substance through non-swelling devices (Cheung, 2013). The extraction yield predicted by this model did not approach to unit with time. The equation is shown in Equation (1):

$$y = Bt^n \tag{1}$$

where y is the yield of *Moringa oleifera* seed oil while, B refer to the constant incorporating the characteristics of the carrier-active agent system, t is the time in minutes and n is the diffusion exponent, an indicative of transport mechanism (Teoh & Mat Don, 2016). The value of n is less than one for the extraction of plant materials. This model is used in the extraction of natural product from different plant which is suitable for *Moringa oleifera.(Louarn et al., 2015)(Louarn et al., 2015)*

Parabolic diffusion model was also used with similar mechanism as the Power Law model but the mechanism is described in two step which consisted of rapid washing step followed by diffusion step (Cheung, 2013). The Parabolic diffusion model is shown in Equation (2):

$$y = y_0 + y_1 t^{\frac{1}{2}} \tag{2}$$

In this equation, the constant y_o shows the extraction yield recovered instantaneous as the *Moringa oleifera* is submerged into solvent (i.e. for $t = 0$) and while y_1 is the diffusion rate constant (Kitanovic et al., 2008).

Naik model applies the time needed to reach half of the extraction yield at an infinite time (Riera et al., 2003). From the engineering point of view, this empirical kinetic model is beneficial to maximize the yield in industrial processes. This model represents the extraction yield of *Moringa oleifera* seed oil as a function of time, t (mins) as Equation (3):

$$Y = \frac{Y_{\infty} t}{b+t} \tag{3}$$

Y_{∞} is the yield after infinite extraction time and while b is the time needed to reach the half of the extraction at infinite time (Riera et al., 2003). Y is the extraction yield of *Moringa oleifera* seeds oil and t is the time needed for the extraction.

The main objective of this study is to investigate the application of UAE to extract oil from *Moringa oleifera* seed by using ethyl acetate as solvent. The influence of these process parameters including temperature, L/S ratio and extraction time were evaluated. The empirical kinetics study on the extraction process will also be studied.

2. Methodology

2.1 Materials

Matured *Moringa oleifera* pods were collected from Kota Tinggi, Johor. The pods were dried for 2 days under the sun before the shells were removed manually. The seeds were grinded and sieved to obtain average diameter of 350 µm. The solvent used for the extraction process was ethyl acetate (QReC), molar weight; 88,10 g/mol.

2.2 Ultrasonic Assisted Extraction of Moringa oleifera Seed Oil

The extraction process of *Moringa oleifera* seed oil was performed in an ultrasonic water bath as shown in Fig. 2, which functioned as indirect ultrasonication. The ultrasonic bath was equipped with a timer, heater, temperature regulator and indicator, as well as ultrasonic power regulator. The ultrasonic bath was set at power of 176 W and ultrasound frequency of 40 kHz. In each run, 4 g of *Moringa oleifera* seeds was mixed with the solvent ethyl acetate at ratio 1:20 otherwise mentioned. The ultrasonic assisted bath was filled with water approximately 2/3 of its volume and instantly immersed the conical flask in the ultrasonic bath. The flask was fixed at the centre of the ultrasonic bath. The extraction process was carried out for a duration, ranging between 0-40 mins. Once completed, the content was centrifuge at 4000 rpm for 20 mins. The solvent in the filtrate was filled in a rotation bottle of rotary evaporator (Heidolph), to obtain the pure oil. The oil yield was calculated using Equation (4):

$$Oil\ Yield\ (\%) = \frac{Mass\ of\ oil\ (g)}{Mass\ of\ seed\ sample\ (g)} \times 100 \tag{4}$$

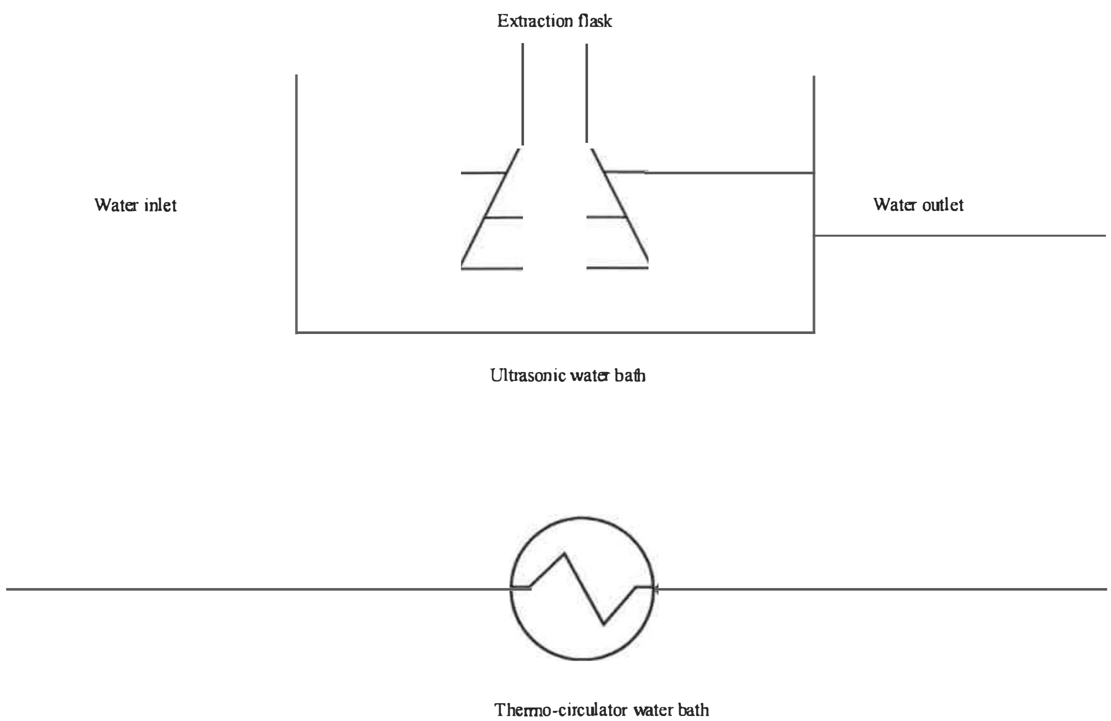

Fig. 2: Schematic diagram of oil extraction using Ultrasonic Assisted Extraction (UAE)

2.3 Empirical Kinetic Model

Table 1: Empirical kinetic model and their linearized form

Empirical model	Equation	Linearized form
Power law model		$\ln y = n \ln t + \ln B$
Parabolic diffusion model	$y = y_0 + y_1 t^{1/2}$	
Naik model	$Y = \frac{Y_\infty t}{(b+t)}$	$\frac{1}{Y} = \left(\frac{b}{y_\infty}\right)\frac{1}{t} + \frac{1}{Y_\infty}$

Empirical kinetic model parameters for all models were estimated using a regression analysis. For each model, the linearized equation was used in the parameter estimation. The proposed models with their respective linearized form equation were shown in Table 1.

3. Results and Discussion

3.1 Effect of Extraction Temperature

The effect of extraction temperature was considered and the data obtained is shown in Fig. 3. The extraction was conducted by using ethyl acetate as the solvent and at different extraction temperature, ranging from 0-60°C and the ratio of solute to solvent was 1:20. It was observed that the oil yield increased as the temperature increases and reached its maximum (35.65%) at 30°C. Higher solubility of the oil in the extracting solvent and a lower solvent viscosity will resulted in higher extraction temperature (Zou et al., 2011). From that, it will 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 (Ramandi et al., 2017). More oil would be extracted into the solvent due to the high heating effect have weaken the cell wall integrity of the seeds and thus provide large surface area between the solvent and the oil (Feng et al., 2015).

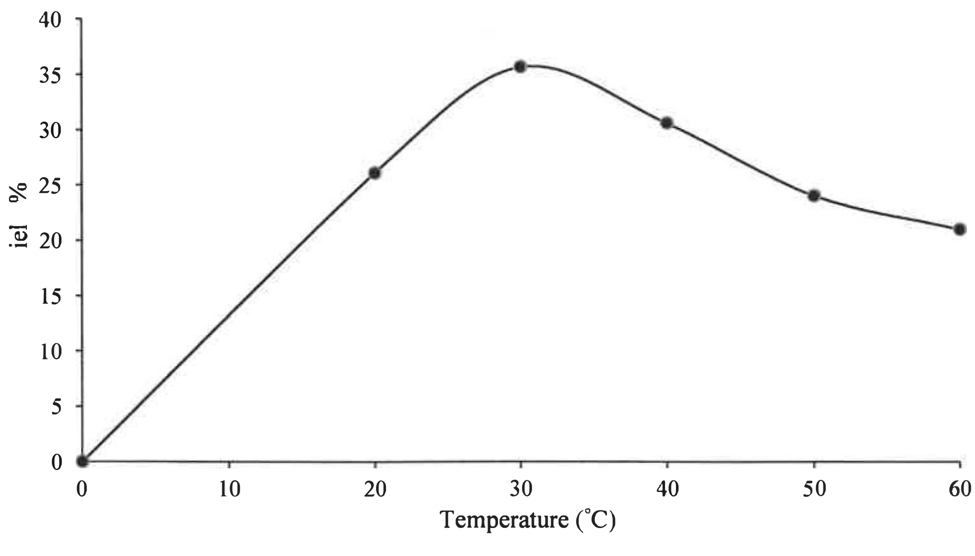

Fig. 3: Effect of extraction temperature on oil yield

Meanwhile, the oil yield did not show any increment after 30˚C. According to Lou et al. (2010), formation of vapor-filled bubbles at elevated temperature and as a result the implosion of cavitation bubbles would be cushioned. This cushioning effect would eventually cause the ultrasound cavitation effect to be less efficient. High vapor pressure of the solvent, the cavitation achieved at the lower ultrasound intensity will reducing the strength of shear forces in the vicinity of the bubbles (Xu et al., 2014). Hence, the decrease an oil yield at higher temperature was expected. The optimum temperature of oil yield was determined to be at 30˚C.

3.2 Effect of Extraction Liquid to Solid Ratio

Fig. 4 shows the effect of different L/S ratio on the percentage of oil yield. The percentages of oil yield were increased as seeds to solvent increased from 1:5 to 1:20. The highest oil yield obtained is 35.14% of *Moringa oleifera* seeds and 80ml of ethyl acetate.

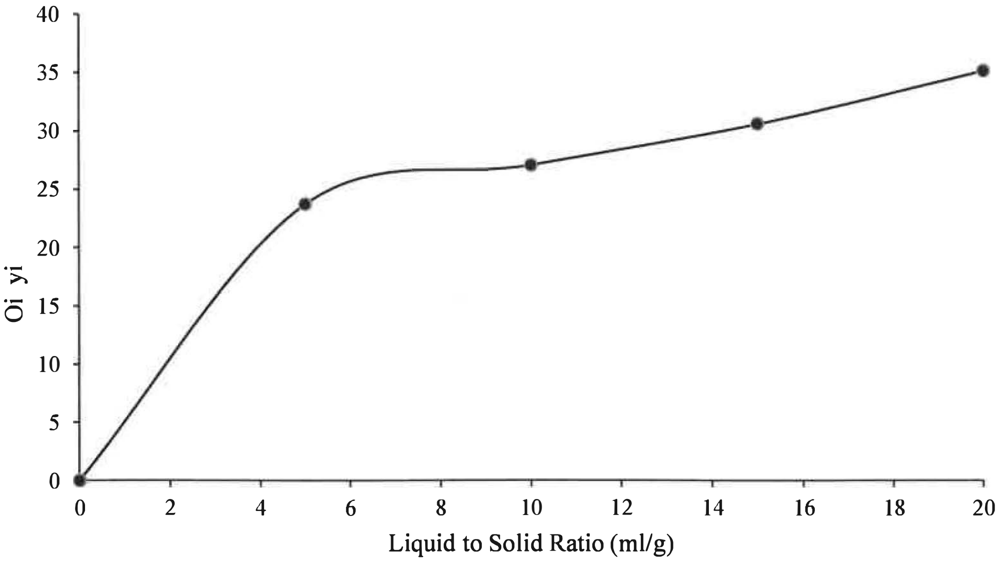

Fig. 4: Effect of liquid to solid ratio on oil yield

These results show that as the volume of the solvent increases, the percentage of oil yield increases. It shows that the volume of solvent has a great effect on extracting oil from the seeds. Thus, the mass transfer driving force between solid and liquid phase has become greater when the solid to solvent ratio increased (Ghafar et al., 2017). Also, as the L/S ratio increased, the extract yield also increased because of the amount of vapour contacted with the seeds increased when the volume of solvent high (Bokhari et al., 2012). This means that further increase of L/S ratio would increase the oil yield and might be significantly unchanged at higher L/S ratio. Hence, the best significant L/S ratio in the UAE of *Moringa oleifera* seed oil was founded to be 20 ml/g.

3.3 Effect of Extraction Time

In this study, the effect of extraction for time on *Moringa oleifera* seed oil extraction is shown in the Fig. 5. The extraction was conducted by using ethyl acetate at different time for about 0-40 mins with constant temperature at 30˚C and the ratio of seeds to solvent was 1:20.