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

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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 176kW power and ultrasonic frequency of 40kHz. Usage of highly toxic and carcinogen solvent was avoided in this study by using ethyl acetate as replacement. The influence of extraction temperature, solid to liquid (S/L) ratio and extraction time were looked at. Experimental data reveals that 38.1% of Moringa oleifera seed oil yield was obtained at temperature of 30^{o}C, S/L ratio of 1:20 and 40 mins of extraction. The study was followed by fitting the experimental data in 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 experimental data with R^2 value of 0.9919 hence, adequate to describe the extraction process of oil from Moringa oleifera seed.

Keywords: *Moringa oleifera*, seed oil, ultrasonic assisted extraction (UAE), empirical model, model fitting, ethyl acetate

I. 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 and 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 utilised as a source of food, as well as in medicinal applications (Mani *et al.*, 2008). *Moringa oleifera* can be cultivated for its leaves, pods or mainly its seed or kernel for oil extraction. The yield varies widely, depending on the season, fertilization and irrigation regimen (Gopalakrishnan *et al.*, 2016). Yielding *Moringa oleifera* seed in 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). Figure 1 shows the Moringa oleifera pods and seeds.

The seed of *Moringa oleifera* is a rich source of oil and protein. It contains oleic acid or 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*

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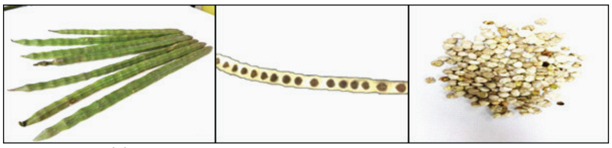
(a) (b) (c)

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

oleifera seed contains fats, fibre, proteins, minerals, vitamins A, B, C as well as also amino acids which are beneficial to human skin (Daba, 2016). In fact, the 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).

To date, there are several established techniques to extract seed oil. They are cold pressing, Soxhlet extraction, and ultrasonic assisted extraction (UAE). Cold pressing is suitable for extraction under mild process conditions (Soto *et al.*, 2007) and this process is applied to extract many other types of seed oil such as *Nigella sativa* (Mohammed *et al.*, 2016) , Chia seed (Ixtaina *et al.*, 2011) and *Calophyllum inophyllum* (Kedzia *et al.*, 2011). This method is highly preferred due to the lower extraction temperature requirement that can avoid oil oxidation in order to maintain its quality (Siger *et al.*, 2017). Meanwhile, Soxhlet extraction offers continuous process for solid-liquid extraction. The extractor consists of thimble, where it contains the material to be extracted. This method uses large amount of organic solvent that is heated in reflux. Unfortunately, this method is capable for only single extraction process and it requires long extraction time, typically 8 hours and the extraction yield is reported to be influenced by the condition of the thimble (Halfadji and Touabet, 2013). The extraction usually occurs at the boiling point of the solvent for a long time. In this case, the possibility of thermal decomposition of the natural substances cannot be ignored.

On the other hand, ultrasonic assisted extraction (UAE) is an alternative 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 and the system involves formation of cavitation that provides shear forces to facilitate the disruption of the cell wall (Sánchez-Madrigal *et al.*, 2017). This condition leads to the transfer of solute into the solvent. In fact, the operation is easy to handle and it requires shorter extraction time as well as lower consumption of solvent (Halfadji and Touabet, 2013).

However, the study on the application of UAE on the oil extraction of *Moringa oleifera* seed is rarely updated, hence its kinetics. 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 where they were normally 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 Power Law Model equation is shown in Equation (1):

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

where y is the oil extraction yield while B refers to the constant by 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 and Mat Don, 2016). The value of n is less than one for the extraction of plant materials.

This model was used in the extraction of natural product from different plant which is suitable for *Moringa oleifera*.

The mechanism of Parabolic diffusion model is described in two steps: rapid washing step and diffusion step (Cheung, 2013) where the equation is as shown in Equation (2):

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

In this equation, the constant y_0 shows the extraction yield recovered instantaneous as the seed is submerged in the solvent (i.e. for $t = 0$) while y_1 is the diffusion rate constant (Kitanovic *et al.*, 2008).

On the other hand, 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 oil as a function of time, t (mins) as shown in Equation (3):

$$y = \frac{y_\infty t}{b + t} \tag{3}$$

where y_∞ is the yield after infinite extraction time while b is the time needed to reach half of the extraction at infinite time.

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 process parameters including temperature, S/L ratio and extraction time on the oil yield were evaluated and the experimental data were used in determining the best fitted kinetic model.

II. MATERIALS AND METHOD

A. 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 350μm. The solvent used for the extraction process was ethyl acetate (QReC).

B. 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 Figure 2, which functioned as indirect ultrasonication. The bath was equipped with a timer, heater, temperature regulator and indicator, as well as ultrasonic power regulator. Ultrasonic power and frequency of 176W and 40kHz, respectively, were used in this study. In each run, 4g of Moringa oleifera seed were mixed with the solvent ethyl acetate at ratio 1:20 in a conical flask, otherwise mentioned. The bath was filled with water approximately 2/3 of its volume to immerse the conical flask together with its content. The flask was fixed at the centre of the ultrasonic bath. The extraction process was carried out for a duration ranging between 0 to 40 mins. Once completed, the content was centrifuged at 4000 rpm for 20 mins. The solvent in the filtrate was evaporated using a rotary evaporator to obtain pure oil. The percentage of oil yield was calculated using Equation (4):

$$Oil\ Yield\ (\%) = \frac{Mass\ of\ oil\ (g)}{Initial\ mass\ of\ seed\ (g)} \times 100 \quad (4)$$

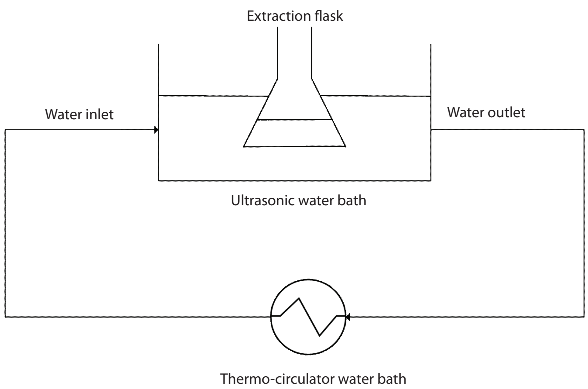

Figure 2. Schematic diagram of oil extraction using Ultrasonic Assisted Extraction (UAE)

C. Empirical Kinetic Model

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

III. RESULTS AND DISCUSSION

A. Effect of Extraction Temperature

The effect of extraction temperature was considered and the data obtained is shown in Figure 3. The extraction was conducted by using ethyl acetate as solvent at different extraction temperature, ranging from 0 to 60^{o}C, whereas the ratio of S/L ratio was kept constant at 1:20. It was observed that the oil yield increased as the temperature increases and reached its maximum (38.1%) at 30^{o}C. Higher extraction temperature resulted in higher solubility of the oil in the extracting solvent as well as low solvent viscosity (Zou *et al.*, 2011). This phenomena contributed to an improvement in terms of mass transfer together with the enhancement of solvent diffusion rate into the pores of the seed (Ramandi *et al.*, 2017). Higher amount of oil is extracted into the solvent due to the heating effect that have weaken the cell wall integrity of the seed, thus providing large surface area for mass transfer (Feng *et al.*, 2015).

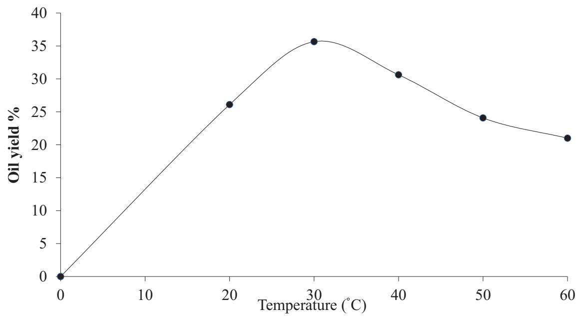

Figure 3. Effect of extraction temperature on oil yield

Further increment of the temperature causes the yield to reduce drastically from 40^{o}C onwards. It is highly possible that this condition is

Table 1. Empirical kinetic model and their linearised form

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

caused by cushioning effect. According to Lou *et al.* (2010), the vapour pressure at elevated temperatures assisted in the formation of vapour-filled bubbles and as a result, the implosion of those bubbles were cushioned. The cavitation effect at high extraction temperature is said to be less efficient, hence has significantly lowered the yield. In this study, 30^{o}C was selected as the best temperature to extract *Moringa oleifera* seed oil.

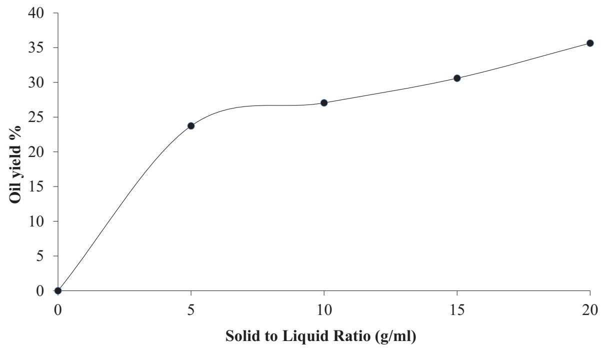

Figure 4. Effect of solid to liquid ratio on oil yield

B. Effect of Extraction Solid to Liquid (S/L) Ratio

Figure 4 shows the effect of different solid to liquid (S/L) ratio on the percentage of oil yield. Based on the data provided, the percentage of oil yield increases together with the increment of solvent volume in the system. It can be observed that the oil yield has increased significantly from 1:5 to 1:20 where the highest oil yield was obtained at S/L ratio of 1:20.

Data in Figure 4 reveals that the volume of solvent has greatly influenced the process of oil extraction from the seed. The mass transfer driving force between solid and liquid phase was enhanced as the S/L ratio increased (Ghafar *et al.*, 2017). Additionally, Faiznur and Abd Karim (2017) reported on the enhancement of mass transfer coefficient for ultrasonic oil extraction during the washing and diffusion stage at high volume of solvent. Though application of greater solvent amount has always been associated with improved extraction, however by considering economical point of view as well as energy requirement, great volume of solvent is reported to be inappropriate. Hence, ratio of 1:20 was chosen as the best condition.

C. Effect of Extraction Time

Moringa oleifera seed oil yield (%) data as function of time is shown in the Figure 5. The extraction was conducted by using ethyl acetate at 30^{o}C and S/L ratio of 1:20 whereas the extraction time was varied from 0 to 40 mins.

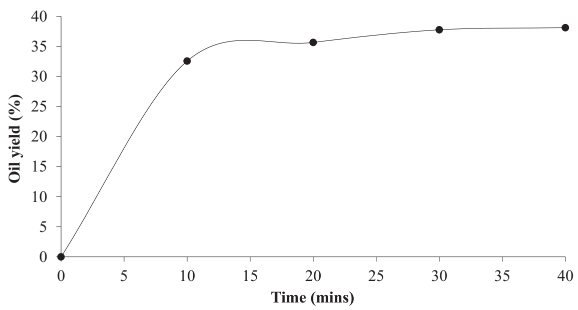

Figure 5. Effect of extraction time on oil yield

Data provided reveals that higher percentage of oil yield was obtained as the extraction time prolongs. The oil yield increases at longer extraction time due to the cracking process on the seed cells. This phenomena happens when it undergoes acoustic cavitation effect during the early stage of extraction (Tian *et al.*, 2013). Best penetration of solvent through the seed cells can be achieved thereby allowing the seed oil to be released into the solvent easily.

However, extending the extraction time beyond 30 mins did not benefit the system as there is no significant increment of the oil yield. The system is said to be in equilibrium state from this point onwards. The oil starts to reabsorb towards the residue of the seed which lowers the yield. As a result, the amount of extractable oil was likely to have reached its maximum level therefore the oil yield remained the same (Faiznur and Mashitah, 2016).

D. Model Fitting

Experimental data obtained were fitted into three empirical kinetic models: Power law model, Parabolic diffusion model, and Naik model. Figure 6, Figure 7 and Figure 8 illustrate the profiles of experimental and predicted data for the *Moringa oleifera* seeds oil extraction using these empirical kinetic models. The experimental data was represented by symbols while the predicted or fitted data was represented by line.

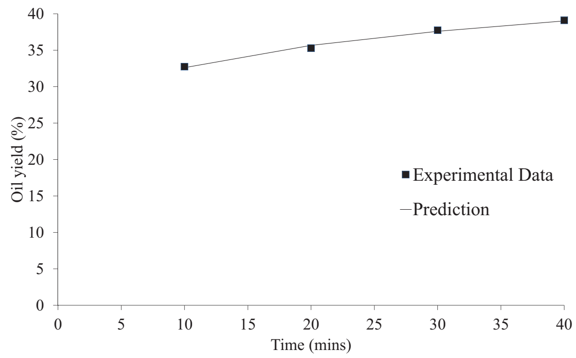

Figure 6. Comparison of experimental and predicted data for ultrasonic extraction of *Moringa oleifera* seeds oil using the Power law model

The predicted results from all three models gave a good agreement to the experimental data, with the coefficient of determination, R^2 ranging from 0.8736 to 0.9919 as shown in Table 2. The lowest R^2 was given by the Parabolic diffusion model which is 0.8736, followed by Naik model (R^2 of 0.9457). Power law model recorded the highest R^2 value (0.9919), indicating that it was the best model to be proposed which describes both, rapid washing stage and diffusion stage for extraction process of *Moringa oleifera* seed oil.

Table 2. Estimated model parameters and correlation coefficient for each empirical model

Empirical model	Model parameter	R^2
Power law model	$B = 24.1963$ $n = 0.1296$	0.9919
Parabolic diffusion model	$y_0 = 4.8769$ $y_1 = 6.1079$	0.8736
Naik model	$y_\infty = 40.9836$ $b = 2.5983$	0.9457

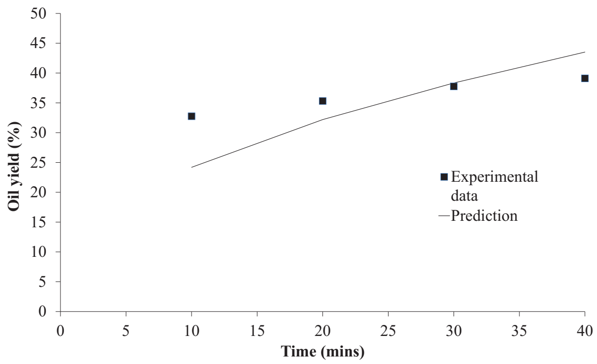

Figure 7. Comparison of experimental and predicted data for ultrasonic extraction of *Moringa oleifera* seeds oil using the Parabolic diffusion model

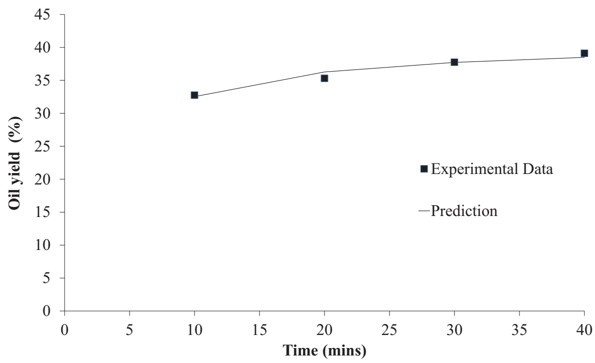

Figure 8. Comparison of experimental and predicted data for ultrasonic extraction of *Moringa oleifera* seeds oil using the Naik model

IV. CONCLUSION

The study explored the extraction of oil from *Moringa oleifera* seed by using ultrasonic assisted extraction (UAE). Ethyl acetate was used as the solvent at temperature and S/L ratio of 30^{o}C and 1:20, respectively. 38.1% of *Moringa oleifera* seed oil yield was recorded in 40 mins. The experimental data was then fitted in three kinetic empirical models and they fitted well with Power law model, at R*2* of 0.9919.

V. ACKNOWLEDGEMENTS

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