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Kinetics of Oil Extraction from Candlenut (*Aleurites moluccana*)

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Oil from candlenut (*Aleurites moluccana*) was extracted using a solvent extraction technique. The influence of three parameters namely extraction time, extraction temperature as well as liquid to solid (L/S) ratio on the candlenut oil yield were studied to optimise the extraction conditions for achieving maximum oil yield. The maximum candlenut oil yield (35.67%) was achieved using methanol as a solvent at a temperature of 45°C for 80 min of extraction period. The optimum L/S ratio was 10ml/g. It was found that the candlenut oil yield increases with the increase of extraction time, extraction temperature and L/S ratio. Kinetics of solvent extraction of oil from candlenut was evaluated using Peleg's model and Logarithmic model. The model parameters were calculated using the experimental data. The kinetics of candlenut oil extraction conforms very well to the Peleg's model with a high R^2 value of 0.9927 and low MRPD value of 1.827%. However, the Logarithmic model can fairly describe the candlenut oil extraction process with the values of R^2 and MRPD of 0.9653 and 4.352%, respectively.

Keywords: *Aleurites moluccana*; candlenut; kinetics; optimisation; solvent extraction

I. INTRODUCTION

Candlenut or *Aleurites moluccana* is a flowering tree that belongs to the Euphorbiaceae family. In Malaysia, it was commonly known as *buah keras* nut tree. Apart from candlenut, *Aleurites moluccana* is also known as Candleberry, Indian Walnut, Varnish tree, Kukui and Kamiri nut tree, depending on the region (Siddique *et al.*, 2011). Candlenut trees are native to the Molucca Islands and widely found in South East Asia (Norulaini *et al.*, 2004). It has been spread throughout the tropical Pacific because of the high oil content in the seeds, which was around 50%. The candlenut oil has been used for thousands of years by Polynesians as a moisturiser since it contains essential fatty acids such as linoleic and linolenic acids that are beneficial to healthy skin. In the pharmaceutical industry, it is used for treating numerous skin diseases such as eczema, psoriasis and burned skin. It is effective in repairing damaged skin. Candlenut oil is also an excellent ingredient in the making of soap, lotion, creams, balms, scrubs or used in massage oils. This candlenut oil is widely used in the cosmetics industry and might currently be candlenut primary commercial product. It is also used as a varnish and in the preservation of fishing nets. Recently, candlenut seed is being exploited as a potential feedstock for biodiesel (Sulistyo *et al.*, 2009).

Isolation of vegetable oil involved several approaches from the ancient time until today. Various methods are used including mechanical pressing, maceration, effleurage extraction, solvent extraction, steam distillation, ultrasonic and supercritical fluid extraction. The method chosen depends on the ease with which the substances can be removed from the plant material, the types of plant material and the costs involved in extracting the substances. Solvent extraction is one of the typical methods of extracting vegetable oil from oilseeds (Ricardo Cardoso *et al.*, 2013). In this method, the solid sample is immersed in a suitable solvent and the extracted oil is collected after the equilibrium extraction is reached (Chan *et al.*, 2014). Generally, solvents such as hexane, ethanol, methanol,

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petroleum ether and acetone are used in the extraction of oil from oilseeds (Mani *et al.*, 2007). Solvent extraction is considered as one of the cheapest and most efficient technique in producing oil from many types of seed such as jojoba, soybean, palm and jatropha (Dutta *et al.*, 2014).

The mathematical kinetic model is powerful engineering tools which greatly facilitate optimisation, simulation and design of an extraction process (Jokić *et al.*, 2010). They are also useful in the development of scaling-up procedures from bench to pilot and industrial scales (Ayas and Yilmaz, 2014). Kinetic models such as power-law and parabolic diffusion models have been used in the extraction study involving numerous target compounds from different types of solid materials (Ghasemi-Fasaei *et al.*, 2012; Das and Bera, 2013; Yaneva *et al.*, 2013). Other than that, second-order model (Kusuma and Mahfud, 2018; Goula, 2013), Sovová model (Piva *et al.*, 2018), Peleg's model (Vetal *et al.*, 2013) as well as Ponomaryov's empirical equation (Veličković *et al.*, 2006) have been widely used in modelling various solid-liquid extraction processes. The semi-empirical kinetic model introduced by Peleg is widely used to explain the extraction curves of biological materials from plant sources because of the similarity to the shape of sorption curves (Vetal *et al.*, 2013). Kinetic modelling provides a theoretical description of the process behaviour and evaluation of the mass transfer coefficients (Simeonov and Koleva, 2012). A model that fits the experimental data is used to determine the diffusion coefficients which are required to describe and optimise the process, thus providing a better understanding of the complex diffusion mechanism (Winitsorn *et al.*, 2008). To date, the evaluation of kinetics on oil extraction from candlenut has not been reported in the literature.

This study aims to investigate the influence of extraction time, extraction temperature and L/S ratio on the extraction yield of candlenut oil. The kinetics behaviour of oil extraction from candlenut was then evaluated using Peleg's model and Logarithmic model. The best-suited model will be determined accordingly. Meanwhile, the experimental data obtained will be used to establish the kinetics of the oil extraction process.

II. MATERIALS AND METHODS

A. Candlenut Oil Extraction

500g of fresh candlenut were purchased from a market located in Shah Alam, Selangor. They were cleaned and stored at ambient temperature. Before oil extraction, the candlenut was ground using a dry mill and sieved through a sieve shaker of pore size 850μm. The sample with particle size of 850μm and below was used in this study. Methanol was used as a solvent. The candlenut sample (5g) and methanol were mixed in a conical flask (125ml), which was then covered with aluminium foil. It is then placed on a hot plate to heat the extraction mixture to the desired temperature. A magnetic stirrer was placed in the extraction mixture to promote mixing. After the extraction process was completed, the oil extract was centrifuged at 4,000rpm 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 (Heidolph). The extracted oil obtained was placed in a desiccator to remove any moisture and weighed until a constant weight was reached. In this study, the extraction time, extraction temperature as well as L/S ratio was varied from 5–120 min, 25°C–55°C and 5–12ml/g, respectively.

B. Candlenut Oil Yield

Extraction yield of candlenut oil was calculated using Equation [1]

$$Y = \frac{M_1}{M_0} \times 100\% \quad (1)$$

where Y is the extraction yield of candlenut oil (%), M_1 is the mass of candlenut oil extracted from the sample (g), and M_0 is the mass of the candlenut sample used (g).

C. Mathematical Model

The kinetic behaviour of the oil extraction process from candlenut was evaluated using Peleg's model and Logarithmic model. The kinetic parameters for both models were calculated numerically with a non-linear regression using Polymath Software 5.0.

D. Peleg's Model

Peleg's model, which is used for the description of sorption curves, was adapted for the description of the candlenut oil extraction process. The expression for Peleg's model is

$$y(t) = y_o + \frac{t}{K_1+K_2 t} \quad (2)$$

where y(t) represents the oil yield at time t, K_1 is Peleg's rate constant (relates to extraction rate at the very beginning of the extraction process), K_2 is Peleg's capacity constant (relates to maximum extraction yield during the extraction process) and y_o is the oil yield at time t = 0. At t = 0, y_o value equals zero, and thus Equation [2] can be reduced to

$$y(t) = \frac{t}{K_1+K_2 t} \quad (3)$$

E. Logarithmic Model

The logarithmic model was used as follows

$$y(t) = a \log t + b \quad (4)$$

where a and b are logarithmic model constants, and $y(t)$ represents the oil yield at time t.

F. Statistical Analysis

The validation of kinetic models was done by evaluating the differences between the experimental and predicted oil yield via determination coefficient, R^2. In addition, the goodness-of-fit of the model was further evaluated using the mean relative percentage deviation (MRPD) value. It is defined in Equation [5]

$$MRPD\ (\%) = \frac{100}{N} \times \sum \frac{|Y - Y_P|}{Y} \quad (5)$$

where Y and Y_P are the experimental and predicted yield of candlenut oil, respectively, and N is the number of experimental data. A mathematical model is considered acceptable to be used in describing an extraction process if the MRPD value is less than 10% (Kaymak-Ertekin and Gedik, 2004).

III. RESULTS AND DISCUSSIONS

A. Effect of Extraction Time

Figure 1 shows the effect of extraction time on the candlenut oil yield at a temperature of 30°C and L/S ratio of 10ml/g. It can be seen that the yield increased rapidly from 14.81% to 29.66% within the first 40 min of extraction period. As the extraction time prolonged until 80 min, the oil yield continues to increase gradually from 29.66% to 32.89%. However, beyond 80 min, the change in oil yield is no longer significant.

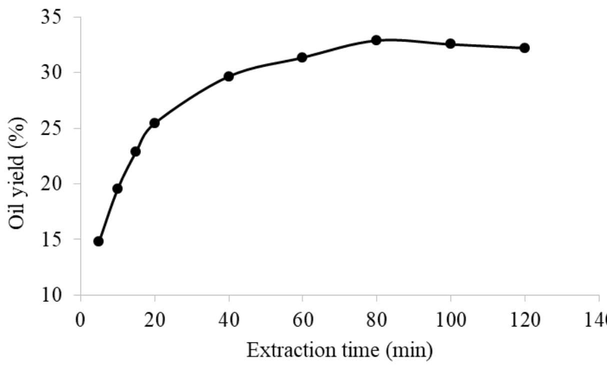

Figure 1. Effect of different extraction time on candlenut oil yield (extraction temperature 30°C, L/S ratio 10ml/g and methanol as a solvent)

Therefore, the optimum extraction time to achieve the maximum yield of candlenut oil (32.89%) was found to be 80 min. According to Goula (2013), this could be because the extraction processes occurred in two stages; the first stage which is characterised by a rapid rate, involved solvent penetration into the cellular structure followed by the dissolution of soluble constituents in the solvent. This is also known as a washing stage. The second stage, better known as a slow diffusion stage, involved the external diffusion of soluble constituents through the porous structure of the residual solid and its transfer from the solution in contact with the particles to the bulk solution. The oil yield does not show any significant changes after 80 min.

A similar result was also obtained by Hu *et al.* (2012) in their study on safflower seed oil extraction. The extraction yield increased with time and reached a maximum value at 30 min. After that, it increased a little. During this stage, the osmotic pressure balanced the solution system and the insignificant increase of extraction yield after a certain extent shows that an equilibrium condition was achieved in the solvent (Norshazila *et al.*, 2017). Therefore, extending the extraction time will not increase the oil yield further.

B. Effect of Extraction Temperature

The effect of extraction temperature towards the candlenut oil yield is shown in Figure 2. During this experiment, both extraction time and L/S ratio were kept constant at 80 min and 10ml/g, respectively. It was observed that the increment of temperature from 25 to 45°C had improved the oil yield from 31.26% to 35.67%, but when the temperature was raised to 55°C, the oil yield dropped gradually to 34.55%.

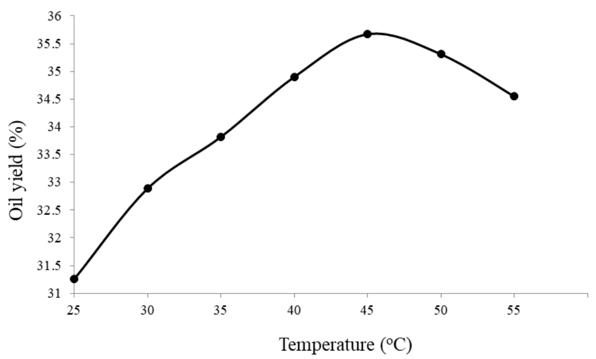

Figure 2. Effect of different temperature on candlenut oil yield (extraction time 80 min, L/S ratio 10ml/g and methanol as a solvent)

This observation can be attributed to the fact that the temperature affected many physical properties such as viscosity, diffusivity, solubility, vapour pressure and surface tension. Liquid with lower viscosity has high diffusivity. Therefore it could easily be diffused into the pores of the solute (Hemwimol *et al.*, 2006). Besides that, the extraction yield was found to increase with increasing temperature due to the increased solubility of the oil in the solvent. In addition, at a higher temperature, the liquid viscosity and density decreased, thus resulting in increased mass transfer (Toma *et al.*, 2001; Palma and Barroso, 2002).

On the other hand, the oil yield started to decrease when the temperature was further raised to 55°C. This might be due to the decomposition of active ingredients presents in the oil (Bhutada *et al.*, 2016). Abdullah *et al.* (2017) reported the same finding in their study on the effect of temperature on the microalgae oil extraction. The extraction temperature was varied over the range of 55⁰C to 75⁰C. The increase in temperature from 55⁰C to 65⁰C leads to an increase in the oil extraction yield from 47.29% to 56.92% while increasing the temperature from 65⁰C to 75⁰C resulted in a decrease in the oil yield from 56.92% to 52.26%. The maximum candlenut oil yield (35.67%) is achieved at an optimum extraction temperature of 45°C.

C. Effect of Liquid-to-Solid Ratio

The relationship between the oil yield and L/S ratio is shown in Figure 3. The extraction temperature and time were kept constant at 45°C and 80 min. The oil yield increases from 31.64% to 35.67% as the L/S ratio increases gradually from 5 to 10ml/g. This increasing trend of oil yield is due to the increasing rate of oil diffusion caused by a higher L/S ratio. A high L/S ratio increases the concentration gradient, and therefore, increases the rate of diffusion. These results are consistent with the mass transfer principles whereby a greater concentration gradient promotes a greater driving force (Norshazila *et al.*, 2017). However, the oil yield did not show any significant increment when the L/S ratio is further raised to 12ml/g. According to Nyamien *et al.* (2015), this might be due to the saturation phenomenon whereby, when the solvent is saturated with bioactive components, the cellular phenomenon of diffusion stops as the equilibrium is achieved. Since the equilibrium is reached at L/S ratio of 10ml/g, the oil yield does not increase after that. A similar trend was observed by Tan *et al.* (2011) in their study on the extraction of antioxidant compounds from *Centella asiatica*. The solid to solvent ratio was manipulated from 1:5 to 1:20, and it was reported that the maximum total phenolic and flavonoid contents were achieved at a solid to solvent ratio of 1:15. In this study, the optimum L/S ratio to obtain a maximum oil yield (35.67%) is 10ml/g.

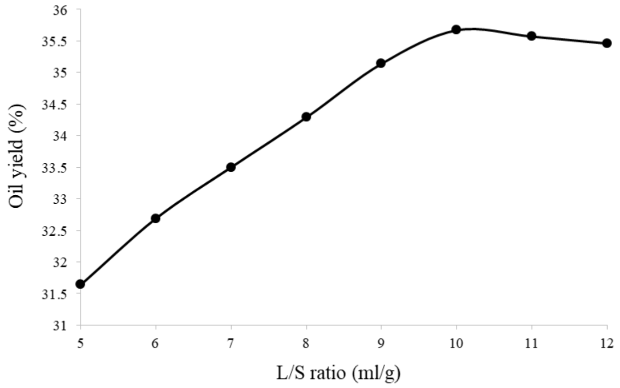

Figure 3. Effect of different L/S ratio on oil yield (extraction temperature 45°C, extraction time 80 min and methanol as a solvent)

D. Kinetics of Candlenut Oil Extraction

The profiles for experimental (represented by symbols) and simulated (represented by lines) oil yield for Peleg's model and Logarithmic model are shown in Figure 4 and Figure 5, respectively. As can be seen, Peleg's model fits the experimental data better than the Logarithmic model. The Peleg's model fit the experimental data in both stages of extraction, namely the washing part and the slow extraction. The washing part occurs at the earlier stage of extraction, from 5 to 40 min while the slow extraction occurs at 40 min of extraction time onwards. The values of kinetic parameters K_1, K_2, a and b are shown in Table 1. In order to validate the kinetic model used, the values of R^2 and MRPD were calculated accordingly. Peleg's model shows a good agreement with the experimental data as the value of R^2 and MRPD are 0.9927 and 1.827%, respectively. These values indicate that Peleg's model is suitable to be used in describing the extraction of candlenut oil. On the other hand, Logarithmic model shows fair agreement with the experimental data obtained as the value of R^2 (0.9603) is slightly lower than that of Peleg's model. Furthermore, MRPD for Logarithmic model (4.352%) is higher than that of Peleg's model.

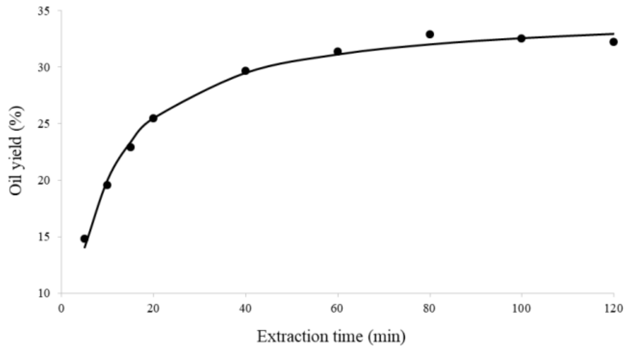

Figure 4. The experimental and simulated oil yield for Peleg's model

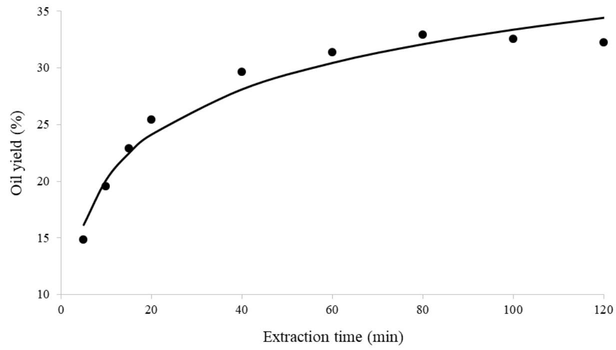

Figure 5. The experimental and simulated oil yield for Logarithmic model

IV. CONCLUSION

In this study, the oil from candlenut was extracted by using the solvent extraction technique. The effects of extraction time, extraction temperature and L/S ratio were evaluated. The oil yield increased with increasing extraction time, extraction temperature, as well as L/S ratio. The kinetics of candlenut oil extraction was studied using Peleg's model and Logarithmic model. The results showed that both models could be used to describe the extraction process with high values of R^2 and low MRPD values. However, Peleg's model fits the experimental data better than the Logarithmic model with a higher value of R^2 (0.9927) and lower value of MRPD (1.827%). Therefore, Peleg's model is more suitable to be used in describing the kinetic behaviour of candlenut oil extraction.

Table 1. Kinetic model parameters for Peleg's and Logarithmic models

Kinetic model	Model parameters		R^2	MRPD (%)
	K_1	K_2		
Peleg's model $y(t) = \frac{t}{K_1 + K_2 t}$	0.213	0.029	0.9927	1.827
	a	b		
Logarithmic model $y(t) = a \log t + b$	13.247	6.887	0.9603	4.352

R^2 : determination coefficient, MRPD : mean relative percentage deviation

V. ACKNOWLEDGEMENTS

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