Kinetic Behaviour and Thermodynamic Study of Peanut Oil Extraction Using Ultrasound Technology

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***Abstract*— In the present study, ultrasound assisted extraction was utilized to extract oil from peanut. There are four type of main operating parameter such as type of solvents, extraction time, extraction temperature and solvent to solid ratio were investigated. The optimum conditions were found at 15 min reaction time, extraction temperature of 40°C, solvent to solid ratio of 20:1 and ethyl acetate as solvent. While, the maximum oil yield is 43.8%. Evaluation of kinetics of oil extraction process was done by using hyperbolic model. High values of coefficient of determination ($R^2 \geq 0.9515$) and low values of the mean relative percentage deviation (MRPD ≤ 0.2759) were obtained, showing the model used were suitable to describe the kinetic of peanut oil extraction. Furthermore, the influence of two parameter of extraction which are extraction temperature and solvent to solid ratio on oil yield were studied. It was found that the oil yield increased with increasing of extraction temperature and solvent to solid ratio. The activation energy (Ea) was calculated as 13.402 kJ mol^{-1} and the extraction of peanut oil was an endothermic process.**

Keywords*— Peanut oil, ultrasonic-assisted extraction, kinetics study, Seed oil

I. INTRODUCTION

Peanut is known as groundnut, while its scientific name is *Arachis hypogaea* (Madhaven, 2001) and it also known as earth nut because of the seeds develop underground. Peanut is cultivated around the world in tropical, sub-subtropical and warm temperate climates (Zhao, Chen et al, 2012). In addition, peanut is very important oilseed in the world where peanut is second ranking of oilseed grown after rapeseed (Jiang, Hua et al, 2010). Generally, peanut contains a massive amount of oil where 40-50% of oil (Li, Jiang, Sui et al , 2011) and 24-36% of protein (Jiang et al., 2010). The fatty acid composition of peanut are influenced by genotype and conditions of growing (Brown, Cater, Mattil et al, 1975) and well demonstrated by (Worthington, Hammons et al 1972).

Peanut is to be popularized in manufacturing of vegetable oil that commonly used in food and cosmetic areas (Xu et al., 2018). According to National Edible Oil Distributor Association, about 22 million ton of production of peanut oil for year of 2017 to fulfil the massive demand of consumers. This is because it contains high nutritional value and delicate fragrance. Generally, peanut oil is packed with omega 3 fatty acids and protein which are responsible for keeping the skin young that can reduce inflammation in the body (Crop, 2001). Peanut oil also can lowers the risk of developing skin cancer. Besides that, it also give hydration to the skin to treat dry and scaly skin (Hashim, Koehler, Eitenmiller et al, 2010).

There are several method of extracting oil from the oilseeds that found by researcher that widely used in the manufacturing scale is solvent extraction. Basically, in traditional industrial production of peanut oil implicate pressing and solvent extraction (Jiang et al., 2010). However, solvent extraction give bad effects such as plant safety problems, emissions of volatile organic compounds into atmosphere, excessive operation expenses, longer processing time and bad quality products caused by using high processing temperatures. Last two decades, some research group recognize new alternative method such as supercritical fluid extraction (Meneses, Caputo et al, 2015), microwave assisted extraction (MAE) (Valdés, Vidal et al, 2015) and ultrasonic assisted extraction (UAE) (Wang et al., 2018) are utilized in extraction oil due to dramatically reduce down working times, growing yields, the high-quality of the extract and recently, these method are identified as an environment friendly extraction method (Jalili, Jafari et al, 2018).

The application of ultrasound technology has been widely studied for extraction oil from the plant materials such as soybean (Cravotto et al., 2008), grape (*Vitis vinifera L.*) seed (Da Porto, Porretto et al, 2013) , rapeseed (Perrier et al., 2017), and pomegranate seed (Tian, Xu, Zhen et al, 2013). The advantages of ultrasound assisted food processing are higher mass and energy transfer ability, lower extraction temperature (Jalili et al., 2018), reduce extraction time, improve rate and oil production (Goula, 2013) and also improve nutritional value, physicochemical and sensorial properties (Mohamed Koubaa, Houcine Mhemdi et al, 2016). Ultrasound had assisted in enhancement of extraction by giving attribution to the propagation of ultrasound pressure waves and resulting cavitation where cavitation bubbles can energetically collapse near the cell wall of the seed and produce localized pressure that causing that cause the plant tissue rupture that give a good penetration of the solvent into the cell, through the ultrasonic jet (Knorr, Ade-Omowaye et al, 2002).

In this work, hyperbolic kinetic model is proposed to represent the kinetics of peanut oil extraction from the determination of kinetic parameters for designing efficient ultrasound extraction process. The implementation of mathematical model of extraction process is very important to ease design of process, to provide information to scale-up the equipment and to optimize and control the extraction process (Bonfigli, Godoy, et al, 2017).

(Zhang et al., 2017) extracted oil from peanut by applying ultrasound technology. However, only the effects of ultrasound power has been studied. Besides that, (S. Haji Heidari, 2017) also studied ultrasound assisted peanut oil extraction but it has addition treatment which is enzymatic treatment with cellulose enzyme. So far, the optimization and kinetic behavior study of ultrasonic assisted peanut oil extraction is not been performed yet.

Thus, the aim of this study were to determine optimize extract parameter of peanut oil using one factor at a time method (OFAT), to evaluate the kinetic of ultrasound assisted extraction (UAE) of peanut oil using mathematical model and lastly, to investigate thermodynamic of the extraction oil process.

II. METHODOLOGY

A. Materials

Peanut was purchased from grocery in Shah Alam. Prior to extraction the impurities of the seeds that consist of stones, broken, and spoiled seeds removed. Without any pre-treatment, the nut were grinded into required measurement which is 0.5-0.75mm through the usage of lab grinder. The sifted seed was stored in sealed bag for further processing Acetone (Merck), ethanol (Merck), methanol (Merck) and ethyl acetate (Merck) were used as a solvent.

B. Ultrasound-assisted Extraction

5 gram of powder peanut were mixed with 50 mL acetone in a 250mL Erlenmeyer flask and immersed into an ultrasonic cleaner bath (Xuba 1) with power of 750 Watt at temperature of 30^{o}C. At the end of extraction, the suspension were centrifuged for 25 min at 6000 rpm. The liquid extraction were put into a rotary evaporator to allow solvent evaporate. The remaining oil was weighed. Four type of solvent were used such as acetone, ethyl acetate, methanol and ethanol. The extraction temperature was manipulated from 30^{o}C to 70^{o}C while the reaction time was varied between 5 min to 30 min. Then, the solvent to solid ratio was investigated from 10:1 – 50:1. These parameters were varied one at a time to determine the optimum conditions for each type of solvent.

C. Determination of Peanut Oil Yield

The yield of oil was determined through the following equations (Wang et al., 2018):

$$\textbf{Oil yield (\%)} = \frac{M_{extract}}{M_{initial}} \times 100 \quad (1)$$

Where:
$M_{extract}$ = Mass of peanut oil extracted from the sample (g).
$M_{initial}$ = Mass of peanut powder sample used (g).

D. Mathematical Model

The hyperbolic model is the one of the kinetic model that used in extraction oil as pelegs model (Menkiti, Agu, & Udeigwe, 2015):

$$q = \frac{C_2 t}{1 + C_2 t} \quad (2)$$

The extraction is first – order at the beginning and decrease to zero – order in the later phase of the process When C_2 t less than 1.

$$q \approx C_1 t \quad (3)$$

And when $t \to \infty$, the equilibrium is reached (q_i –q_e). So,

$$q_e = \frac{q_e}{q_o} = \frac{C_2}{C_1} \quad (4)$$

Equation (3.5) is obtained when hyperbolic kinetic model equation is linearized.

$$\frac{1}{q} = \frac{1}{C_1} \times \frac{1}{t} + \frac{C_2}{C_1} \quad (5)$$

The plot of 1/q that is 1/oil yield against 1/t gives slopes as $1/C_1$ and intercepts as C_2/C_1 whereas C_1 and C_2 are hyperbolic model parameters which are extraction rate at the beginning (min^{-1}) and constant related to maximum extraction yield (min^{-1}), respectively.

E. Statistical Analysis

The model performance had been evaluated through calculating the mean relative percentage deviation (MRPD) value (Fuad & Karim, 2017) and the coefficient of correlation R^2 (Kammoun Bejar, Boudhrioua Mihoubi et al, 2012) between the experimental and the hyperbolic kinetic model (predicted data) as the following equation :

$$\textbf{MRPD (\%)} = \frac{100}{N} \times \sum \frac{|Y - Y_p|}{Y} \quad (6)$$

$$R^2 = \frac{\Sigma(Y_p - Y_{mean})^2}{\Sigma(Y_{exp} - Y_{mean})^2} \quad (7)$$

Where N can be defined as the total number of experimental data. Y and Yp are yield of peanut oil for experimental and prediction, respectively. If the percentage value of MRPD is less than 10%, the mathematical model is considered suitable to use in describing extraction oil process.

F. Calculation of activation energy

The relation between of rate of constant and extraction temperature can be expressed by the Arrhenius Equation as following (Subroto, Manurung et al, 2015) :

$$k = A \exp \left[\frac{-E}{RT}\right] \quad (8)$$

Where:
k = the rate constant of extraction (min^{-1}).
A = the temperature independent factor (min^{-1}).
E = the activation energy (J mol^{-1}).
R = Gas constant (8.314 J mol^{-1} K^{-1}).
T = Absolute temperature in K.

III. RESULTS AND DISCUSSION

A. Effects of solvent.

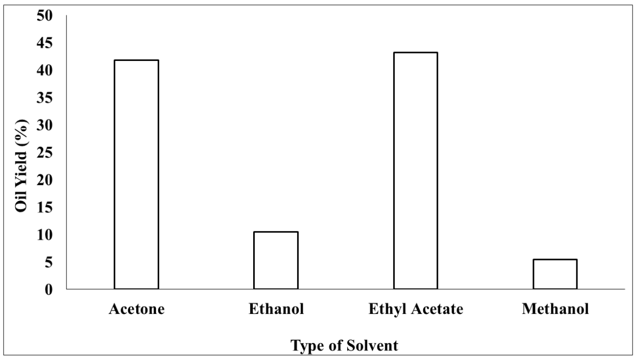

Figure 1: Effect of solvent on oil yield using ultrasound technology with S/S of 50 mL/g for 30 min.

The extraction yield with ethyl acetate was found to be the highest from others (43.2%). Then, it was followed by acetone (40.3%), ethanol (10.5%) and then methanol (5.4%) under similar conditions. According to (Tian et al., 2013) reported that the efficiency of ethyl acetate as solvent for the pomegranate seed oil extraction is higher. The polarity of the solvent is the one of the factor that influenced extraction yield (Silva, Garcia et al, 2016). Based on the polarity index (Sadek, 2002), ethyl acetate is slightly less polar than acetone, ethanol and methanol where the value of polarity indexes of ethyl acetate, acetone, ethanol and methanol are 4.4, 5.1, 5.2 and 5.1, respectively. So, it can be said that ethyl acetate extracted more oil than others. According to (A. Abdolshahi, M.H. Majd et al, 2015), ethyl acetate is the is known to be a good solvent for extracting unsaturated fatty acid. For the other studies from (Silva, Garcia et al, 2016) reported that the performance of ethyl acetate as solvent for chia seed oil extraction gave the highest oil yield than isopropanol.

B. Effects of extraction time

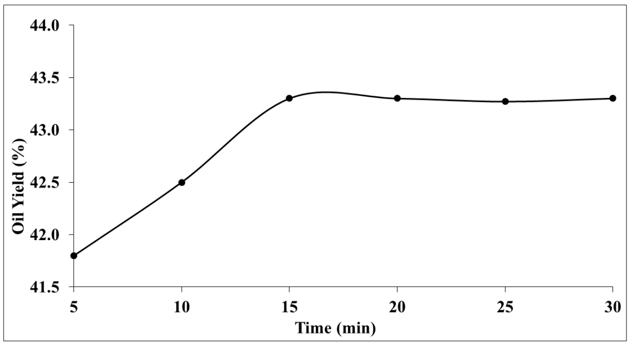

Figure 2: The effect of reaction time on oil yield that using solvent of ethyl acetate with S/S ratio of 50mL/g at temperature of 40°C that assisted by ultrasound technology.

The effect of extraction time on oil yield was shown in Figure 3. This figure show the comparison of extraction with ethyl acetate as solvent, S/S ratio of 50mL/g for different treatment time from 5 to 30 min with steps size of 5 min. From the graph, oil yield increased significantly in the initial 15 min from 41.8% to 43.3%. After 15 min, oil yield reached an equilibrium and significantly decrease to 43.2% until 30 min. All peanut cell wall cracked completely within the first 15 min from the acoustic cavitation effect, leading to good penetration of the solvent into the cell (S. Hemwimol, P. Pavasant, 2006) and enhancing the transfer of dissolved oil out of the solid structure (M. Palma, 2002). As the time increased, the cell wall of peanut was ruptured that resulting to the lowering the solvent's permeability into cell structures. In addition, target components also re-adsorb into the ruptured tissue particles due to their relatively large specific surface areas, lowering oil yield (L. Paniwnyk, E. Beaufoy et al, 2001) A similar trend has also been suggested by (Liu et al., 2017) and (Perrier et al., 2017).

C. Effect of extraction temperature

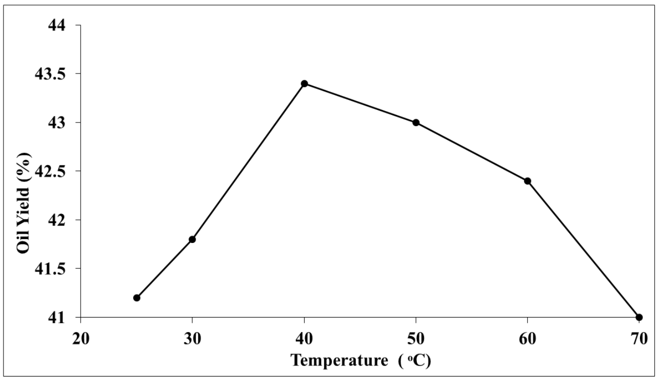

Figure 3: The effect of reaction temperature on oil yield that using solvent of ethyl acetate with S/S ratio of 50mL/g for 15 min that assisted by ultrasound technology.

A range of temperature treatment which are from 25°C to 70°C were employed for 15 min assisted by ultrasonic sound with S/S ratio of 50 mL/g to assess their capabilities on the extraction oil from peanut. Result from the graph show the yield of oil increased rapidly as temperature increase from temperature of 25°C to 40°C (41.2% to 43.4%). However, when the temperature exceeded 40°C, the oil yield decrease significantly from 43.0% to 41.0%. As temperature increased, it will attribute to the increase solubility of peanut oil in solvent which is ethyl acetate. Moreover, when the temperature exceed the optimum temperature, the oil yield will become decrease significantly due to the increasing of mass transfer was resulted from the decreasing of viscosity and density of solvent (Z.S. Zhang, L.J. Wang et al, 2008) (Shalmashi, 2009). The number of cavitation bubbles within the fluid increased creating a cohesive force reducing the tensile strength of the liquid as a result of decreased viscosity of solvent (M. Palma, 2002) (M. Toma, M. Vinatoru et al, 2001). The same order of efficiency was also obtained by (Tian et al., 2013)

D. Effect of solvent to solid ratio

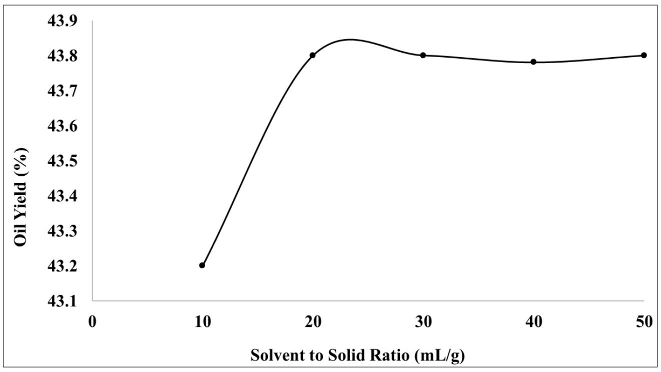

Figure 4: Effect of solvent to solid ratio on the yield of oil at temperature of 40°C for 15 min.

Five solvent to solid ratios of the effects of using ethyl acetate on oil yield were studied including 10, 20, 30, 40 and 50 ml/g at 40°C for 15 min, to determine the capabilities of the ratio of solvent to solid on oil extraction. Graph of Figure 4 shows the oil yield increase from 43.2 to 43.8% with the ratio of solvent to solid increasing from 10 to 20 ml/g. However, when the solvent to solid ratio exceeded 20 ml/g, the oil yield is not significantly increase, a larger amount of solvent will not change the driving force (S.S. Herodez, M. Hadolin et al, 2003). With increasing of the solvent to solid ratio, the oil concentration between the extraction liquid and the materials also increase rapidly due to the effect of rate of diffusion that led to an enhancing of oil yield (Q.-A. Zhang, Z.-Q. Zhang et al, 2009). However, the excessive of solvent would give the effect of constant due to requirement of more energy and time to reflux the extraction solution. Hence, the optimum cycles for extraction could not be reached. Similar resulted had also been obtained by (Sayyar, Abidin et al, 2009) and (Tian et al., 2013)

E. Kinetic study of oil extraction

1) Effect of extraction temperature

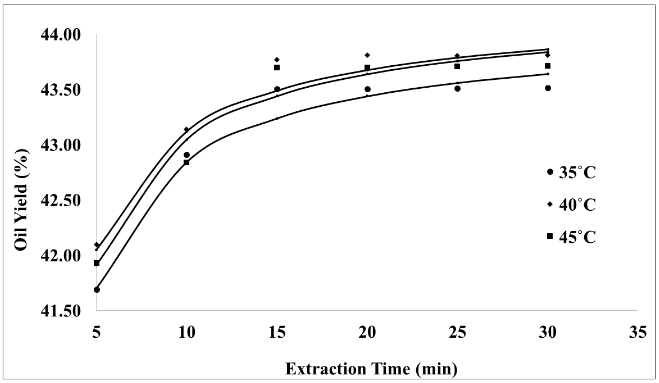

Figure 5: Hyperbolic model kinetic of Peanut oil at different temperature using solvent of ethyl acetate, solvent to solid ratio of 20:1 for 30 min.

Table 1: Extraction rate and maximum extraction yield rate for different extraction temperature

Extraction temperature (°C)	Extraction rate, C_1 (min^{-1})	Maximum extraction yield rate, C_2 (min^{-1})	MRPD (%)	R^2
35	1.5625	0.03547	0.2243	0.9883
40	1.6949	0.03831	0.2022	0.9826
45	1.5873	0.03587	0.2759	0.9589

In the peanut oil extraction that assisted by ultrasound technology, it can be seen that the oil yield of peanut was increased rapidly at the beginning and then, reached equilibrium value with the length of extraction time. Hyperbolic kinetic model was applied to determine and study the kinetic model on the peanut oil extraction that assisted by ultrasound technology. To study the hyperbolic kinetic model on the extraction of the peanut oil, it can be done by plotting the graph between 1/q and 1/t. The slope and intercept from plotted graph were used to determine value of extraction rate, C_1 and maximum extraction yield rate, C_2. From the result obtained, extraction rate, C_1 were 1.5625, 1.6949 and 1.5873 min^{-1} for extraction temperature of 35°C, 40°C and 45°C, respectively.

Basically, the increment of extraction rate C_1 and maximum extraction yield rate C_2 occurred with the increasing extraction temperature as shown in Table 1. However, when temperature exceed to 40°C, the value of extraction rate decreased significantly due to the massive amount of bubbles were formed (Parthiban & Perumalsamy, 2016). From that, they had collapsed with low of intensity. Moreover, when temperature is too high, it can caused the decreasing surface tension of the extracting solvent and automatically, it will affect the formation of bubbles and collapse (Nwabanne, 2012).

From the Table 1, it can be seen that hyperbolic kinetic model for peanut oil extraction had coefficient of determination (R^2) of 0.9883, 0.9826 and 0.9589 for extraction temperature of 35,40, 45°C, respectively. While, the value of MRPD were 0.2243, 0.2022 and 0.2759 for temperature of 35°C, 40 °C and 45 °C, respectively. Hence, it can be said that hyperbolic kinetic model is able to present well the experimental results of peanut oil. This is also been supported by (Menkiti et al., 2015) which states that hyperbolic kinetic model reasonably described TC kernel oil extraction.

2) Effect of solvent to solid ratio

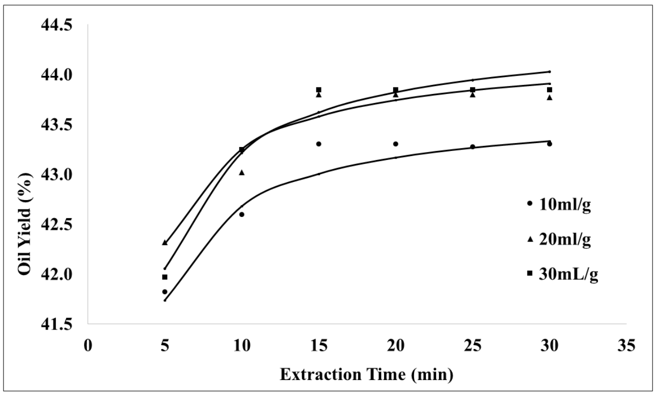

Figure 6: Hyperbolic model kinetic of Peanut oil at different solvent to solid ratio using ethyl acetate as solvent with temperature of 40 °C for 30 min.

Table 2: Extraction rate and maximum extraction yield rate for different solvent to solid ratio (mL/g)

Solvent to solid ratio (mL/g)	Extraction rate, C_1 (min^{-1})	Maximum extraction yield rate, C_2 (min^{-1})	MRPD (%)	R^2
10	1.8868	0.04321	0.2500	0.9770
20	1.9231	0.04346	0.2337	0.9515
30	1.5625	0.03516	0.2467	0.9656

The effect of solvent to solid ratio (mL/g) was studied between 10:1 to 30:1 at constant of extraction temperature 40°C. From the result obtained in Figure 6 shows that the oil yield increased rapidly with solvent to solid ratio at the early beginning of the process from 1 min until 15 min and then the oil yield remain constant after 15 min of extraction time. The extraction rate of the hyperbolic kinetic model were determined from the data of experimental are shown in Table 2. From the result obtained, extraction rate, C_1 were 1.8868, 1.9231 and 1.5625 min^{-1} for solvent to solid ratio of 10mL/g, 20mL/g and 30mL/g, respectively.

An increasing solvent to solid ratio from 10:1 to 20:1 with increasing the extraction rate, C_1 and maximum extraction yield rate, C_2. However, when the solvent to solid ratio exceed 30:1, the extraction rate, C_1 and maximum extraction yield rate, C_2 decrease significantly. As the solvent to solid ratio increases, the gradient of concentration between solute and solvent is higher. Hence the mass transfer increases. Once there is a sufficient amount of solvent available for extraction, further addition of solvent does not affect the extraction yield (Charpe & Rathod, 2016). This is also supported by (Lin et al., 2012) which stated the excessive solvent would not increase the driving force effectiveness due to the limitation of the oil transfer is more confined to the solid interior.

The values of determination coefficient (R^2) for solvent to solid ratio of 10:1, 20:1 and 30:1 are 0.9767, 0.9515, and 0.9656, respectively. The higher value of the determination coefficient indicated that experimental data fit the model very well. While, the value of MRPD for solvent to solid ratio of 10mL/g, 20mL/g and 30mL/g are 0.2500, 02337 and 0.2467. It shows that the model is reasonably to be used in describing the extraction of peanut oil.

3) Calculation of activation energy.

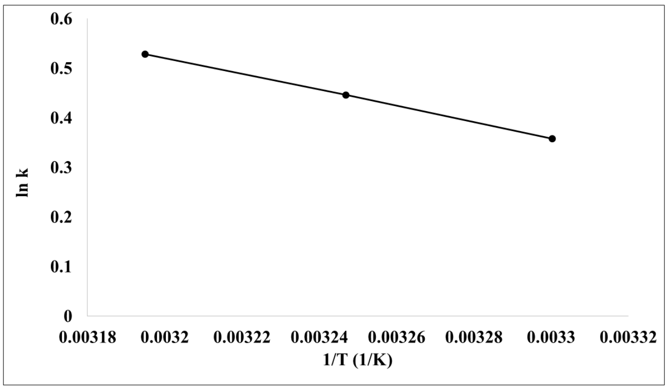

Figure 7: Linear relationship between extraction rate constant of hyperbolic model, ln (k) and temperature for peanut oil extraction.

The extraction rate of hyperbolic kinetic model increase with the extraction temperature as shown in Table 1 and the changes can be described by the Arrhenius Equation. From the Figure 7, it shows a linear representation of Arrhenius Equation. The relationship between k and T can be defined by the linearized Arrhenius Equation (Eq. 3.3), where A and activation energy were found to be 292.5396 min^{-1} and 13402.17 J mol^{-1} respectively obtained from the intercept and the slope of the plotted graph.

IV. Conclusion

There are four main type of operating parameter that influenced the peanut oil extraction that assisted by ultrasonic technology. The optimum condition for the solid liquid extraction from peanut in lab scale were obtained at temperature of 40^{o}C, extraction time of 15 min, solvent to solid ration of 20:1 and ethyl acetate as the solvent used. Ethylene acetate was the most effective solvent for oil extraction followed by acetone, ethanol, and methanol. The hyperbolic kinetic model can represent well the experimental results of peanut oil extraction by ultrasound technology. The experimental data agrees well with the hyperbolic kinetic model and kinetic parameters namely the extraction rate, C_1 and maximum extraction yield rate C_2 with the determination coefficient in range of 0.95- 0.96 and low the value of MRPD. The activation energy have been calculated using the model where the value of Ea is 13402.17 J mol^{-1}.

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