OPTIMIZATION OF THE PROCESSING PARAMETER ON MICROWAVE ASSISTED EXTRACTION CUM STERILIZATION BY USING BOX BEHKEN DESIGN

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***Abstract*—In this study, the optimization of free fatty acid and energy efficiency of palm fruit using microwave assisted extraction was conducted. The objective of this study are to obtain optimum condition for percentage of energy efficient and percentage of free fatty acid by manipulating three process variable which are mass, time , power and effects of the variables towards both response. A set of experiments was design by using Box Behken Design was used to achieve the objectives. The experimental data then fitted to second-order polynomial and analyzed by analysis of variance (ANOVA) using design expert software. Based on ANOVA, R^2 was 0.9585 for percentage of energy efficiency and 0.9284 for percentage of free fatty acid and percentage of energy efficiency was depend on mass as compared to time and power. The three dimensional (3d) response surface plots demonstrate the effect of independent variables on two different generated response .The higher percentage of ffa and percentage of energy efficiency was depend on mass as compared to time and power. An experimental test had been conducted to compare the optimum value predicted by the software. The error for the percentage of ffa and percentage of energy efficiency were 0.0016 and 0.2618.**

Keywords*— *Free Fatty acid, ANOVA, Box Behken, Energy efficiency, Palm Oil

I. INTRODUCTION

Malaysia is one of the biggest country that exported palm oil in the world. Malaysia Palm Oil Industry (MPOB) had reported that Malaysia cover 12% and 27% of total export in the world for palm oil and fats. As a key player in palm oil industry, Malaysia play important role to fulfill global demand for oil and fats. The process of palm oil production consist of few important phase in order to produce high quality of crude palm oil. Palm oil process involve of bunch reception from the plantation, bunch sterilization and threshing, digesting and pulp pressing out of crude palm oil. The crude palm oil is then purify and dry before transport to refinery.

Bunch sterilization is one of the crucial process in palm oil extraction. . Sterilization process is a process of heat treatment to protect the quality of palm oil by deactivate lipase on producing free fatty acid(FFA)(1). Sterilization process also help to loosening and separate palm fruit from the bunch. Fresh fruit bunches (FFB) are sterilized using steam at temperature range 140˚C to 150˚C for period 75-90 minute. A study indicate that longer time with high temperature cause of sterilization in microwave might result lower content of free fatty acid concentration as more lipase activity is deactivate (2).

Conventional drying method such as hot air drying still can be categorized as preservation technique but it affects final quality of dried product in term of nutrient, color and shrinkage. Microwave radiation is a new alternative method in drying application whereby heat is provide to the whole material volume in lesser time as compared to conventional drying method. A study have proved in their study that processing of fresh palm fruit by using microwave that microwave treatment able to replace conventional sterilization method in order to destroy enzymatic lipase in palm oil, fruit softening and nut conditioning (3). Microwave method have high potential in developing continuous, dry and clean technology in palm oil mill process. High power microwave also heats product more efficiently compared to lower microwave (4). But too high power might cause rapid increase in temperature (temperature runaway) that may reduce the quality of product (5)

Research surface methodology (RSM) comprises a group of mathematical and statistical technique used in development of relationship between independent variable, x and generated response, y.Basically, the relationship between independent variable and generated response is unknown. Hence, the relationship is determine by approximate the function ()For experimental data perform in curvature, the data can be calculate using second degree model or known as second order polynomial model. Three experimental data that usually used which are Box Behken Design, Doehlert Design, Central Composite Design .By applying RSM, model equation can be identify and verify by using analysis of variance (ANOVA) function. ANOVA represent key idea in statistical modelling of complex data structure which the grouping of predictor variables and their coefficient into batches (6).The coefficient determination ($R^{2)}$ value generated by ANOVA will defined the ability of the model. ANOVA will come out with several in order to verify the equation. The main objective of response surface methodology (RSM) is to improve the quality a response model including reduction of the variable, process quality and product performance (7). Other than that, this method is able to reduced number of trial experiment in order to evaluate multiple variables and their generated response. This study, the optimization of percentage of free fatty acid and percentage energy efficiency with assistance of microwave radiation is carried out. A set of experimental were designed by box-behken design using response surface methodology. Mass, time and power will be manipulated as to see the effect om percentage of free fatty acid and percentage of energy efficiency.

METHODOLOGY

A. Microwave Assisted Extraction cum Sterilization (MAECS)

The microwave oven used for organic solvent free microwave assisted extraction *cum* sterilization (MAECS) was a household microwave Panasonic, model: NN-ST651M, 1000 Watt frequency of 2450 MHz. In order to examine the effect of microwave power level, 800 Watt to 100 Watt were studied. The dimensions of the interior cavity of the oven were 35.5 cm (W) 25.1 cm (H) and 35.6 cm (D). Microwave oven was modified by drilling a hole at the top(8). Flat bottom quartz flask having a capacity of 1000 ml was placed in the cavity and connected to condenser through the hole(9)(10) as illustrates in Figure 1. Temperature was monitored by a shielded thermocouple K-type (SE-305) inserted directly into the quartz flask. After the condenser was placed near the microwave, the hole around the neck of the flask was covered with polytetrafluoroethylene (PTFE) to prevent leakage of microwave. It is therefore pretty important that this radiation does not escape the microwave and start zapping people standing nearby.

Prior to extraction oil method, only several spikelet of bunch were selected and weighed before undergoing microwave heating for 2 min intervals time until 100% stripping reach. The trials were repeated 3 times for 400, 600 and 800W of microwave power level. The fruits at the spikelets were detached with a slight push of a finger [12, 16] until the fruits were completely detached from the spikelet. The stripping efficiency was then determined according to Eq. (1).

$$\%\ Stripping\ Efficiency, S_{eff} = \frac{F_l}{F_l + F_r} x100\% \quad (1)$$

where S_{eff}, the percentage of stripping efficiency, F_l, the number of fruits loosened from the spikelet after stripping, and F_r, the number of fruits remained at the spikelet after stripping. In addition, study is necessary on a combined system by using combination of moisture and heat in a microwave so that sterilisation can be conducted at shorter time with 100% fruits stripping efficiency. Effect of water ratio (m:v) at 1:0.5, 1:1 and 1:2 was also investigated for each batch of microwave revealing duration, where the ratio is fruits mass (gram) to water volume (ml). As the exposure duration was extended, there was significant increase in drying, cracking and browning effect on the mesocarp and kernel. This step is essential to give them the initial moisture prior to sterilization process (8). The condition and appearance of the center of the kernel was noted.

Figure 1 indicates the design rig for extraction of palm oil via organic solvent free microwave extraction *cum* sterilization. The moistened plant material as indicates in Figure 2, was placed in flat bottom quartz flask connected to cooling system apparatus. During the process, the vapour passed through the condenser outside the microwave cavity where it was condensed. The extraction process was performed for different times and was continued until no more possesses a sweet fragrant aqueous was obtained. For each condition, experiments were replicated twice. The aqueous was collected in vials, dehydrated with anhydrous sodium sulphate, capped under nitrogen and kept at 4 C until being analysed. Parameter interest is as explained in next section.

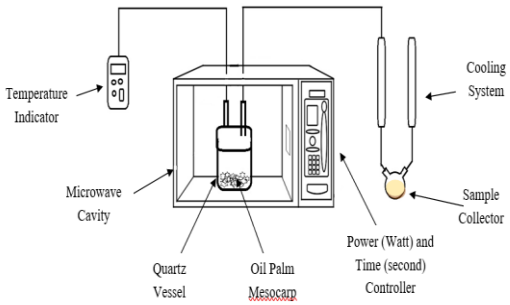

Figure 1: Schematic diagram of organic solvent free microwave assisted extraction cum sterilization (MAECS)

Figure 2: Manually peeled of mesocarp

B Post Treatment of Oil Extraction

This section will be described the post treatment for output from quartz vessel after MAECS process. After irradiated by MAECS, oil palm mesocarp in the quartz vessel was rapidly cooled down to 40 °C in an ice bath, with solvent to feed ratio of 1g/g, 100 ml of absolute hexane was added to the sample and the experiment proceed with leaching and constantly stirred using magnetic stirrer for 80 min to homogenizing the temperature of the mixture during the extraction process. The method was adapted from (11). Afterwards, extracts were subjected to rotary vacuum evaporation (Heidolph Laborata 4000) at 40 °C to remove the solvent from the extract palm oil. The evaporated product obtained under different conditions were analysed by gas chromatography (GC Varian 450-GC). The aqueous extract was clarified using separating funnel where it was left to stand for 60 min at a constant room temperature to form two distinguished layers of methyl ester - solvent and aqueous. The clear separated methyl ester layer was dried with anhydrous sodium sulphate prior to injection into gas chromatography for analysis.

C Free Fatty Acid & Energy Efficiency

Free fatty acid was determined by using titration method. Sample was pippete until the sample turn to reddish purple colour while penolptelain act as indicator. Volume of 0.1 NaOH was measured and calculated by using equation (2).

$$Free\ fatty\ acid, \% = \frac{MWx\ Vx\ n}{w} \quad (2)$$

Where MW is molecular weight, V is volume and n is 0.1N for NaOH normality and w is weight of sample.

Energy efficiency defined as an objective to reduce amount of energy required. The energy efficiency calculated by using equation (3)

$$Energy\ effciency, \% = \frac{Energy\ output}{energy\ input} x100 \quad (3)$$

D Box Behken Design

In order to obtain optimum condition for free fatty acid and energy efficiency, a sets of experiment were conducted by manipulating three process variable which time, power and mass. The quadratic equation model for predicting optimal point is expressed in equation (4).

$$y = ß_0 + \sum_{i=1}^{k} ß_1 + \sum_{i<j} \sum ß_{ij} x_i x_j + \sum_{i=1}^{k} ß_{ii} x_i^2 + € \quad (4)$$

Where Y is the response, $ß_0$ is constant coefficient, $ß_1$, $ß_{ij}$ and $ß_{ii}$ are coefficient for the linear, quadratic and interaction effect $x_i and\ x_j$ are independent variables while € is the error.

The Design Expert (Version 6.0.8) was used in order to obtain regression and graphical analysis based on the experimental data. In this design, Response Surface Methodology act as method to calculate optimum value in the software. The experimental data was fitted into second-order polynomial and analyzed by analysis of variance (ANOVA) using design expert software. The variability in dependent of variable was determined by value of R^2 and the quadratic equation (4)to predict the optimum values.

II. RESULTS AND DISCUSSION

A. Experimental design

The data from the experiment will be analysed in quadratic model. Two quadratic regression models were made by using actual value from data in table 1. The model for equation 1 was developed by generated response for percentage of energy efficiency while equation 2 was for free fatty acid content. Both models develop based on independent variables power, time and mass.

$$Energy\ Efficiency\ \%, Y_1 = 31.33 - 11.08A - 7.28B + 4.76C + 4.41A^2 - 3.61B^2 - 2.61C^2 - 3.91AB - 2.78AC - 1.33BC \quad (2)$$

$$FFA\ \%, Y_2 = 0.31 - 0.016A + 0.003175B + 0.029C - 0.002948A^2 + 0.007052B^2 + 0.018C^2 + 0.007275B - 0.020AC + 0.018BC \quad (3)$$

Run	Actual Value			Free fatty acid content (FFA) %	Energy efficiency %
	A, Power (W)	B, Time (Min)	C, Mass (g)	Actual	Actual
1	100	5	100	0.3392	51.49
2	400	10	120	0.2893	25.6
3	100	10	120	0.3238	30.84
4	250	15	80	0.3030	20.59
5	250	100	10	0.2970	38.73
6	250	5	120	0.3110	18.03
7	250	10	100	0.3392	51.49
8	400	10	120	0.3110	22.00
9	250	5.00	80	0.3072	27.2
10	250	15	80	0.2867	13.84
11	250	5	120	0.3482	39.35
12	250	15	120	0.3968	20.39
13	250	10	100	0.3059	33.67
14	250	10	100	0.325	27.86
15	250	10	100	0.3059	33.67
16	250	10	100	0.3059	33.67
17	250	10	100	0.3059	33.67

Table 1: Free Fatty Acid based on palmitic acid

B. Analysis of Variance (ANOVA)

ANOVA is analysis of variance which used to evaluate the model fitted quality. ANOVA is a software with statistical procedure used to analyse degree of two or more vary in an experiment. The relationship between independent variable and generated response is analysed in order to obtain significant result. The generated response was corresponding to the second order polynomial fitting data. In this research, three independent variables were power, time and mass while generated response were energy efficiency and free fatty acid content.

Based on Table 2, ANOVA for response surface method for analysis of energy efficiency was obtain. The model can be consider as significant model by looking to the F value and prob>F. In this model, the F-value was 17.98 and prob>F was 0.0005 which indicate the model was significant. A model can be categorized as significant whenever the value of F was higher while prob>F was small. The model F value was 1.32 which implies the model insignificant on lack of fit relative to pure error. There was 38.58% chance that a ‘’lack of fit F value’’ occur due noise. Non-significant lack of fit was good as it show that the model was fit. The value of predicted R-squared of 0.6380 was not as close as adjusted R-squared of 0.9052. This may indicate a large block effect or a possible problem with the model or data. The value of R^2 was 0.9585 which higher that adjusted R^2 indicate the model was adequate.

Source	Sum of square	DF	Mean square	F-Value	Prob>F
Model	1845.5	9	205.06	17.98	$5x10^{-4}$
Residual	79.82	7	11.40		
Lack of fit	39.63	3	13.21	1.32	0.3859
Pure error	40.19	4	10.05		
Correction Total	1925.5	16			
R^2	0.9585	Adj R^2	0.9052	Pred R^2	0.6380

Table 2: ANOVA for Response Surface Quadratic Method for Analysis of Energy Efficiency.

The data of ANOVA for response surface method for analysis of free fatty acid content was show in table 4.3. The model can be consider as significant model by looking to the F value and prob>F. In this model, the F value was 10.09 and prob>F was 0.003 which indicate the model was significant. There was only 0.3% chance that a ‘’model F-value’’ this large occur due to noise. The model F value was 3.45 which implied the model insignificant on lack of fit relative to pure error. There was 13.14% chance that a ‘’lack of fit F value’’ this large occur due noise. Non-significant lack of fit was good as it show that the model was fit. The value of predicted R-squared of 0.1431 was not as close as adjusted R-squared of 0.8364. This may indicate a large block effect or a possible problem with the model or data. The value of R^2 was 0.9284 which higher that adjusted R^2 indicate the model was adequate.

Source	Sum of square	DF	Mean square	F-Value	Prob>F
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Lack of fit	39.63	3	13.21	1.32	0.3859
Pure error	40.19	4	10.05		
Correction Total	1925.5	16			
R^2	0.9284	Adj R^2	0.8364	Pred R^2	0.1431

Table 3: ANOVA for Response Surface Quadratic Method for Analysis of free fatty acid.

C. Three dimension (3D)

The three dimensional (3d) response surface plots demonstrate the effect of independent variables on two different generated response which percentage of ffa and percentage of energy efficiency in figure 1. Figure 3.a shows that time and power were two manipulated variable that resulted in quadratic effect on free fatty acid percentage. The percentage of free fatty acid depends more on time compared to power. This is because as the time increasing, the microwave heating also increase. The increment of free fatty acid was due to the destruction of micro structural barrier in the kernel and endocarp by heat generated during microwave treatment and increase the formation of lauric acid (12). Figure 3.b which mass and power revealed quadratic effect on percentage of free fatty acid. The percentage of free fatty acid was depends more on the mass compared to power. The higher mass of palm fruits the higher content of free fatty acid. Thus, as palm fruit sterilized in the microwave, higher percentage of free fatty acid will remain in the palm fruit compared to lower mass of palm fruit. In fact, the lower the free fatty acid content the higher the quality of palm oil. The similar situation is shown in figure 3.c whereby two manipulated variable mass and time contribute significant effect on percentage of free fatty acid. Based on three dimensional (3d) of figure 3.c, it show that percentage of free fatty acid slightly depends to the mass as compared to the time.

Figure 3.d shows the quadratic effect on percentage of energy efficiency after the reaction between power and time illustrated in the 3d response surface plot. The energy seem to be constant and start to decrease as the time increase while energy efficiency was high at low power and start to decrease and kept constant as the power increase. Based on the graph it shows that percentage of energy efficiency was depends more on the time rather than power. This is because the higher level of power usage in microwave will cause a waste on energy consumption. Figure 3.e shows the quadratic effect of power and mass on percentage of efficiency. As can be seen in figure 3.e, an increment of energy efficiency as the mass increase while percentage of energy efficiency was constant as the power increase. Figure e show similar situation as figure 4f whereby percentage of energy efficiency could not depend on power because an increase in power may cause a waste on energy if it not consume properly. Figured 3.f show that the percentage of energy efficiency was rely on mass rather than time. This is because as the mass of palm fruit increase, the energy was consumed to heat the palm fruit. Thus, no energy been wasted.

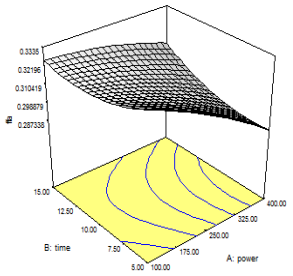
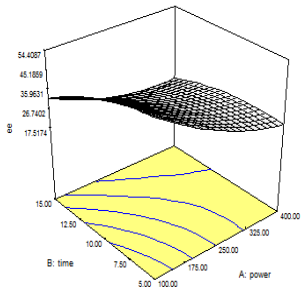
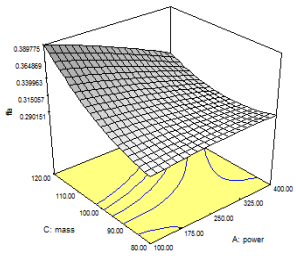
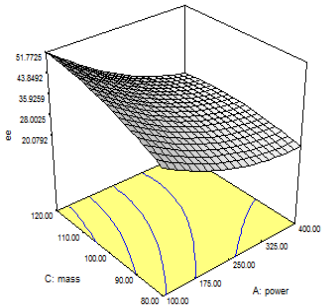
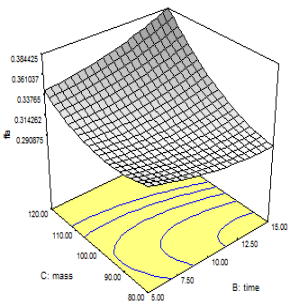
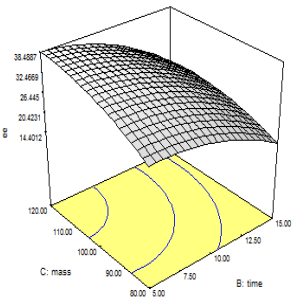
Free Fatty acid%(FFA)	Energy efficiency(EE)
a.  X=B:Power Y= C:Time A:Mass =100	d.  X=B:Power Y= C:Time A:Mass =100
b.  X=B:Power Y= C:Mass A:Time =10	e.  X=B:Power Y= C:Mass A:Time =10
C.  X=B:Time Y= C:Mass A:Power =250	f.  X=B:Time Y= C:Mass A:Power =250

Figure 3:Response Surface 3D for effect of parameters on energy efficiency and free fatty acid.

D. Optimisation

Optimization of percentage of free fatty acid and percentage of energy efficiency in microwave sterilization of palm fruit was done in order to achieve desired criteria of the response model. The percentage of free fatty acid to be minimize while percentage of energy efficiency was set maximize. All the independent variable was kept in range. The percentage of free fatty acid was set 0.2857 to 0.3958 and percentage of energy efficiency was set 13.54 to 53.83.

Based on desirability function, the software will come out with five best solution of independent variable to achieve optimum condition of generated response. The optimizations show that the highest desirability was 0.803 but it generated three solution. Since only one best solution to be picked up, the optimum combination of independent variable that developed best response will be chosen.

E. Validation Model

Based on optimum condition obtain from optimization, an experiment was perform in order to validate the optimum condition. Table 4.5 show the value for predicted from the optimization and experimental value.
Table 4.5: Model validation

	A	B	C	Predicted	Actual
1	100	7.63	91.42	0.3116	0.3100

Table 4: Model validation for percentage free fatty acid

	A	B	C	Predicted	Actual
1	100	7.63	91.42	47.3382	47.6000

Table 5: Model validation for percentage Energy efficiency

The value for predicted and experimental for percentage free fatty acid was 0.3116 and 0.3100. The percentage error was 0.0016. Besides, for percentage of energy efficiency the value for predicted was 47.3382 and experimental value was 47.600. The percentage error between predicted and experimental was 0.2618. Since the value was close, the model can be used in optimizing of palm fruit for percentage of free fatty acid and percentage of energy efficiency using microwave-assisted extraction cum sterilization (MAECS) .

III. Conclusion

The objective of this study is to achieve optimum condition for two variables which are percentage of free fatty acid and percentage of energy efficiency towards by using on Microwave-assisted extraction by using RSM. The optimization of the processing parameter design by using box-behken design and calculated by secondary order polynomial model. The analysis for percentage of free fatty acid showed that the F-value was 10.09 and prob<F was 0.003 which indicate the model suitable to be use. Whilst, F-value and prob<F for percentage energy efficiency were 17.98 and 0.005.The value was significant for a design model. Both generated response also show non-significant lack of fit which indicate the model well-fitted. Other than that, both model show highest R^2 where 0.9585 for percentage of energy efficiency and 0.9284 percentage of free fatty acid. The value of adjusted R^2 for both design were 0.9052 for percentage of energy efficiency and 0.8364 percentage of free fatty acid. Lower value of adjusted R^2 rather than R^2 show that model is adequate. The three dimensional (3d) response surface plots demonstrate the effect of independent variables on two different generated response .The higher percentage of ffa and percentage of energy efficiency was depend on mass as compared to time and power. An experimental test had been conducted to compare the optimum value predicted by the software. The error for the percentage of ffa and percentage of energy efficiency were 0.0016 and 0.2618. The lower error implies that the model can be used in optimizing of palm fruit for percentage of free fatty acid and percentage of energy efficiency using microwave-assisted extraction cum sterilization (MAECS).

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