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Full Length Research Paper

Demulsification of water-in-oil (W/O) emulsion via microwave irradiation: An optimization

Abdurahman H. Nour[1]*, A. N. Ilia Anisa[1] and Azhari H. Nour[2]

[1]Faculty of Chemical Engineering and Natural Resources, University Malaysia Pahang, Malaysia.
[2]Faculty of Industrial Sciences and Technology, University Malaysia Pahang, Malaysia.

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The formation of water-in-crude oil emulsion creates many problems in petroleum industry such as decreasing the oil recovery efficiency, high pumping cost, and piping corrosion, which needs to be treated before processing. Microwave irradiation was employed to demulsify the water-in-oil (W/O) emulsion, which encountered in refinery industries. Three types of crude oils consisted of heavy and light crude oil used in this study to compare the effectiveness of microwave and conventional heating. The optimal conditions for microwave irradiation were determined by response surface methodology (RSM) for each crude oil. Correlation analysis of the mathematical regression models indicated that quadratic model could be employed to optimize the microwave irradiation in each crude oil. The optimal conditions for crude oil A were microwave power of 714 W, time processing in microwave irradiation of 2.53 min with 0.15 wt% of demulsifier and the percentage yield of water separation was 45.00%. While for crude oil B, the maximum yield of water separation predicted through this model was 64.07% at 692 W in 2.56 min. In crude oil C, the maximum yield of water separation predicted through this model is 38.03% at microwave power 767 W in 2.50 min with 0.14 wt% demulsifier.

Key words: Demulsification, water-in-oil (W/O) emulsion, optimisation, response surface methodology.

INTRODUCTION

Demulsification is a process of breaking emulsion, which is necessary in many applications such as environmental technology, painting, petroleum industry and waste-water treatments. There are two types of demulsification approach in refinery industry, which are physical and chemical methods (Yang et al., 2009). It has been known for a long time that microwaves are used to heat materials. In fact, the development of the microwave oven for the heating food has been more than a 50-year history (Wu, 2003). Recently, microwave is investigated as an alternative method to break the emulsion. The concept of microwave irradiation in demulsification was introduced by Klaika (1978) and Wolf (1986). Microwave irradiation offers clean and convenient heating process and in most times has a better result in percentage of water separation due to the rapid heating with uniformity (Abdurahman et al., 2007; Rajaković and Skala, 2006). Phenomena in microwave irradiation involved with electromagnetic waves interact within the materials or molecules.

The heating of liquids using microwaves can be explained by the interaction of matter with the electric field of the incident radiation, causing movement of ions as well as that of induced or permanent molecular dipoles. This movement can cause heat generation. In microwave heating, the most important thing is volumetric heating which in a manner different with conventional heating. Volumetric heating means that materials can absorb microwave energy directly and convert it into a heat. As reported by Nadkarni (1984), for conventional heating, typical time required to complete heat wet absorption by conductive heating is about 1 to 2 h. While for microwave irradiation, the time required is less about 5 to 15 min, which is 96% less than conventional. In

*Corresponding author. E-mail: nour2000_99@yahoo.com.

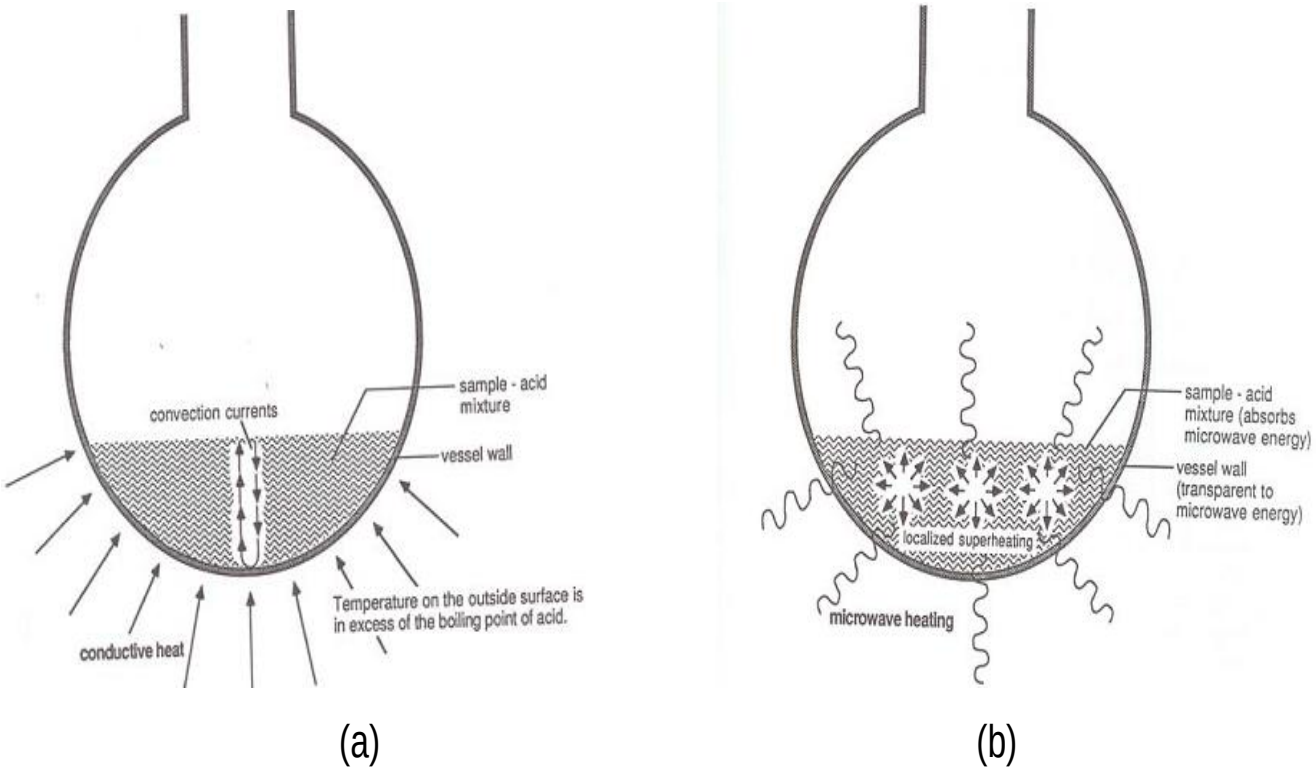

Figure 1. Sample heating by (a) conventional thermal heating (b) microwave heating (Kingston, 1988).

conventional heating, the vessel used usually poor conductors of heat, thus it takes time to heat vessel and transfer into the sample. Because vaporization occurs at the surface of liquid, a thermal gradient is established by convection currents and only a small portion of solution is at the temperature of the heat applied. Therefore, when conductively heating occurs, only a small portion of the solution is above the boiling point temperature of the solution (Figure 1).

The optimization of microwave irradiation for demulsification process implies determining of the experiment's conditions for separation of water from crude oil. In the preliminary study, the demulsification of each crude oil in microwave irradiation was based on the types of crude oil. For heavy crude oil (A and C), microwave-assisted chemical, system might be used to separate the emulsion instead of using only microwave irradiation. Meanwhile for crude oil B, emulsion could be separated in microwave irradiation without being assisted by chemical demulsifier. Thus, the optimization of demulsification in microwave irradiation is significant to improving the percentage of water separated and reducing the dosage of chemical demulsifier used in heavy crude oil.

The present study was conducted with the following objectives: (1) to compare the demulsification of W/O emulsion in microwave irradiation and conventional heating; (2) to design the demulsification in microwave irradiation experiments using central composite design (CCD) and optimize the independent variables using response surface methodology (RSM). The effectiveness of W/O emulsion in microwave irradiation and conventional heating were compared by observing the percentage of water separated, and droplets size distribution in each crude oil.

MATERIALS AND METHODS

Materials

This study was conducted in 2010 at UMP laboratory; the types of crude oil used are; crude oil A, B and C, respectively. These crude oils were donated from Petronas Penapisan Melaka, Malaysia. Each crude oil was characterized physical and chemical. The characterizations are listed in Tables 1 and 2, respectively:

Emulsion preparation

Basically, emulsion is prepared by adding water to crude oil in the prescribed ratio. About 50 ml of emulsion sample was prepared in this study. The prepared water-in-oil (W/O) emulsion formed with the original crude oil were found to be extremely stable and there was no separation observed (under gravity) even after a few days. Firstly, 0.1% emulsifier of crude oil added to crude oil (mixing solution), and was stirred using three blades propeller for 1 min with ±500 rpm. Water was added gradually to the mixing solution and was agitated vigorously using a standard three blade propeller at

Table 1. Physical properties of crude oil.

Variable	Crude oil A	Crude oil B	Crude oil C
viscosity (cP) at 25°C	183.6	24.6	207.8
density (g/cm^3)	0.8459	0.8345	0.8494
^{o}API density	29.226	33.819	26.481
pour point (°C)	-20.4	-19	-12
Wax Appearance Temperature (WAT),°C	-1.18	3.11	4.44
Water content (%)	7	0.65	2
Surface tension (m/Nm)	13.276	13.046	13.659

Table 2. Chemical properties of crude oil.

Types of crude oil	Asphaltenes (A)	Resin (R)	Oil		Wax	Solid particles	R/A
			Non-volatile	Volatile			
Crude oil A	11.00	21.60	33.49	16.97	2.74	14.20	2.70
Crude oil B	5.94	32.33	45.51	0.32	5.11	9.77	11.0
Crude oil C	14.7	23.80	29.01	5.28	11.81	15.27	2.03

1500 rpm and at temperature 28 to 30°C for 5 min. The prepared emulsion was checked whether W/O or oil-in-water (O/W) using test tube and only W/O emulsion was selected for further steps. The concentrations of water (internal phase) in the samples were varied by volume. Emulsions were observed over a period of time to provide a qualitative measure of the stability (Figure 2).

Microwave irradiation in demulsification

The demulsification of water-in-oil (W/O) emulsion was conducted using a domestic microwave oven; Elba domestic microwave oven model: EMO 808 SS. 100 ml of emulsion was inserted into a glass beaker before covered at the top and bottom glass beaker with aluminium foil and was placed in the centre of the microwave. Three thermocouples were inserted in the emulsion samples at different locations; top, middle, and bottom, as shown in Figure 3, respectively. The emulsion samples were heated with microwave radiation at 2450 MHz for a different microwave exposure time. Temperature profiles of emulsions inside the cylindrical container during the batch microwave heating were recorded by a Pico-TC-08 data logger. Meanwhile for conventional heating, hotplate was used in breaking the prepared emulsion. Same volume of prepared emulsion was added into a beaker before heating with hotplate for 3 min. In each minute, temperature of prepared emulsion was measured using a thermometer.

Response surface methodology

Response surface methodology (RSM) was developed by Box and collaborators since 50's and one of the methodologies in determining the optimum results (Kalavathy et al., 2009; Bezerra et al., 2008). RSM is a statistical technique for designing experiments, building models, evaluating the effects of various factors. Moreover, this RSM is useful in finding the optimum values for each studied variable (Lenth, 2009; Sawela and Lele, 2009, Wang et al., 2008, Montgomery, 2005). In this study to evaluate curvature from an optimal graph, a second-order model must be used. A model for a second-order interaction presents the following terms:

$$y = \beta_o + \sum_{i=1}^{k} \beta_i x_i + \sum_{i=1}^{k} \beta_{ii} x_i^{\,2} + \sum_{i} \sum_{\substack{j \\ i<j}} \beta_{ij} x_i x_j + \varepsilon \qquad (1)$$

Where y represents the measured response and x_i, the value of factors. β_o, β_i, and β_{ij} are the constants representing the intercept, coefficients of the first-order term and coefficients of second-order interaction terms and ε is the residual associated to the experiments, respectively.

Design of experiment using central composite design

The software Design Expert (Version 6.0.8, Stat-Ease Inc., Minneapolis, MN) was employed for experimental design, data analysis and modelling of experiment. The central composite design (CCD) of response surface methodology was used to obtain data that fits a full second order polynomial (Sawale and Lele, 2009; Wang et al., 2008). Five replicates at the centre of the design were used to allow for estimation the pure of sum squares (Wang et al., 2008). In this study, two types of factorial design selected based on the types of crude oil. For crude oil A and C, 2^3 factorial design was used which three variables engrossed; microwave power (X_1), time processing (X_2) and concentration of demulsifier (X_3). Meanwhile in crude oil B the design involved was 2^2 factorial design with microwave power (X_1) and time processing as variables (X_2). Each independent variable had 3 levels which were -1, 0 and +1. The distinct of factorial design were summarised in Tables 3 and 4.

Evaluation of the fitted model

The mathematical model found after fitting the function to the data sometimes not satisfactorily described the experimental domain studied. Thus, the model fitted was evaluated using the Analysis of Variance (ANOVA) obtained from Design of Expert. The analysis based on variance ratios was to determine whether significant or not significant difference exist among the means of observed

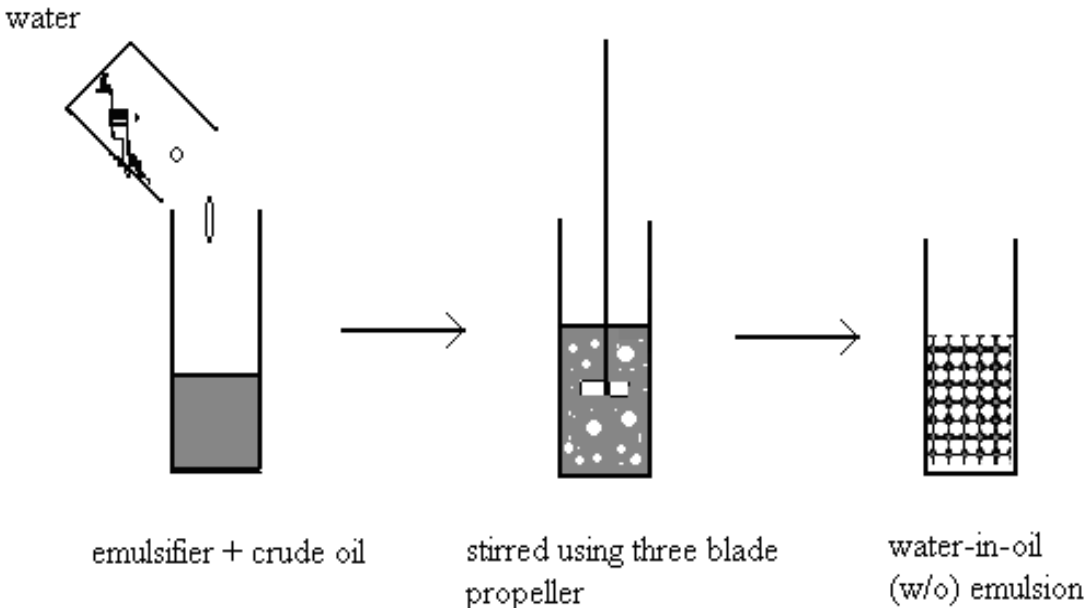

Figure 2. Schematic diagram for preparing W/O emulsion system.

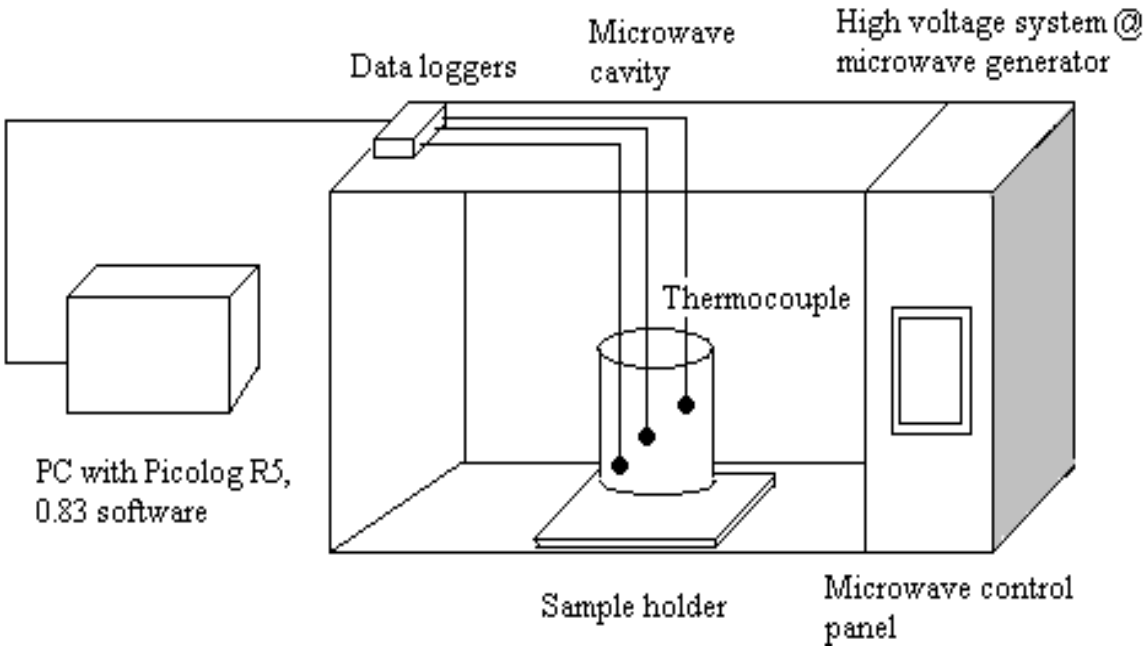

Figure 3. Experimental apparatus used for microwave irradiation demulsification.

parameters.

The analysis has begun with the estimation the effect of each experimental factor and their two factors interaction by estimation the regression and standard error for each coefficient. The significance of each coefficient was determined using the p-value. Another way to evaluate the model is the lack of fit (Bezerra et al., 2008). A model will be well fitted to the experimental data if it presents a significant regression and non-significant lack-of-fit.

RESULTS AND DISCUSSION

Comparison between microwave and conventional heating

The fundamental in different methods of transferring

Table 3. Levels of the experimental variables in 2^3 factorial designs.

Level of experiment variables				
Experiment variables	**α = -1**	**α = 0**	**α = +1**	**Units**
Power	540	720	900	Watt
Time	2	2.5	3	Min
Concentration	0.05	0.1	0.15	wt %

Table 4. Levels of the experimental variables in 2^2 factorial design.

Level of experiment variables				
Experiment variables	**α = -1**	**α = 0**	**α = +1**	**Units**
Power	540	720	900	Watt
Time	2	2.5	3	Min

energy from the source to the sample is the main benefit of utilizing the microwave irradiation as compared to conventional heating process. The energy delivered to microwave-absorbing material is directly penetrated into the sample. The delivering energy generates heat by collision's dipole moment molecules in the emulsion. Thus, the viscosities of microwave and conventional heating can be examined due to the heat-transfer mechanisms from both methods.

The viscosities of conventional and microwave irradiation for 50 to 50% W/O and 20 to 80% W/O emulsions are shown in Figure 6 to 9. Both ratios showed the lowest viscosity obtained once the emulsion was demulsified in microwave irradiation. Indeed, the percentage of viscosity for 50 to 50% W/O emulsion in both before and after being demulsified in microwave irradiation were higher than conventional method for crude oils A, B and C were 87.3, 91.4 and 89% while for conventional, the percentage decreasing viscosity in crude oil A, B and C were 7.64, 32.34 and 20.4%. This was because of the mechanisms of heating emulsion in both methods that was different. In conventional heating, heat was applied to the surface of a body, stimulating the outside molecules or atoms. The kinetic energy is gradually transferred to the inside molecules or atoms until the entire body is heated. While microwave irradiation allows volumetric heating of samples by penetrated the electromagnetic wave (Mutyala et al., 2010).

The lower ratio (20 to 80% W/O emulsion) also displayed the same result which, viscosity of emulsion was reduced after demulsified in microwave irradiation. However, the decreasing viscosity was lesser than the higher ratio because of the content of droplets in 20 to 80% W/O emulsion was lower than 50 to 50% W/O emulsion. The effect of viscosity was related to temperature. At the lower phase ratio, the continuous phase content was dominant thus temperature cannot distribute easily through the whole body of sample in conventional heating. In microwave irradiation, the lower phase ratio reduced the efficiency of electromagnetic to heat emulsion, which waters only was selected and has the ability to absorb electromagnetic field (Figure 4).

The droplets size distribution of the crude oils before and after demulsified the emulsion were illustrated in Figures 5 and 6. Figure 5 indicates that the scatter of droplets size in crude oil B is higher as compared to crude oil A and C due to the lower viscosity of crude oil thus; droplets can interact with each other easily as compared to crude oil A and C. While when compared with crude oil A and C, crude oil C illustrated non-statistical distribution due to the sedimentation and weak interaction between the droplets. This figure supported the crude oil characteristics listed in Tables 1 and 2. The droplets size distribution can be concluded generally to become much tighter as the droplet's size is increased.

After emulsions were demulsified in microwave irradiation, the droplets was literally high and scattered far away from each other and the droplet size distribution was quite uniform. The quite long distance between droplets existed in crude oil B. However, the size distribution in crude oil A and C does not display high scattered droplets because of the high viscosity of the crude oil itself, and this was reasoned for the lower separating of emulsion even demulsified in the microwave.

The effectiveness of microwave irradiation was also determined by observing the droplet's diameter of emulsion.

According to Figure 6, the emulsion droplets were dependent on the effectiveness of demulsification process in microwave irradiation. As discussed previously, the emulsion does not separate in microwave irradiation, and demulsifier was added to enhance the separation process. This happens to all emulsions except the emulsion prepared from crude oil B. The left side (a, c

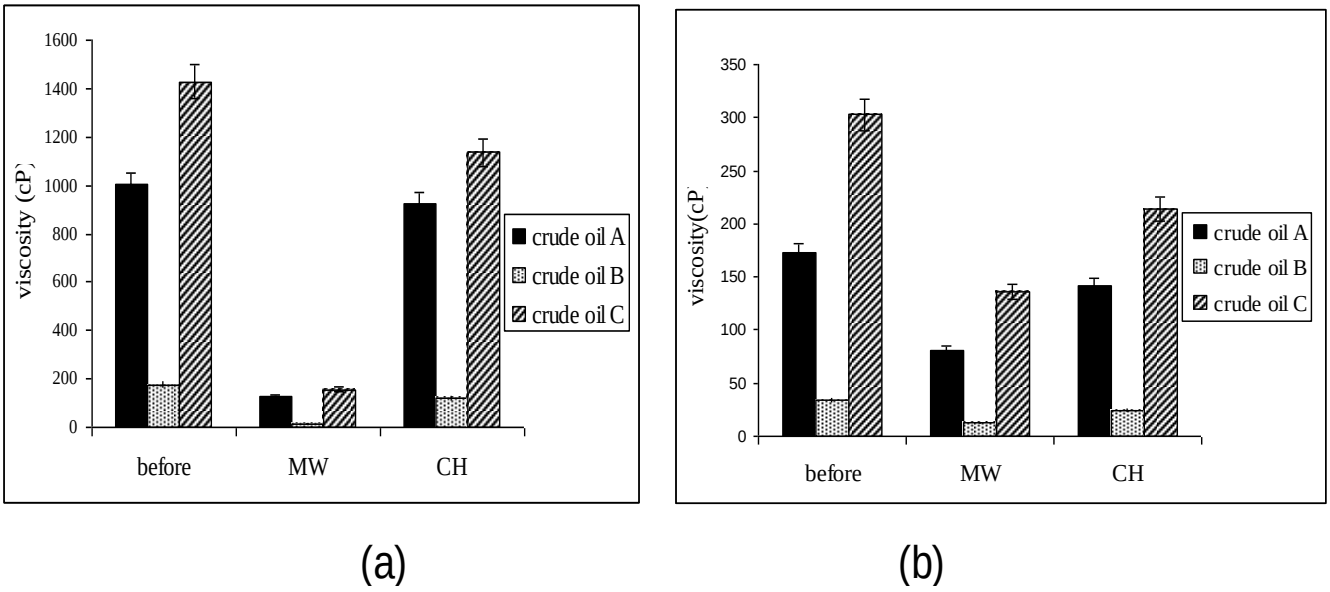

Figure 4. Effect of viscosity by comparison microwave (MW) and conventional heating (CH) (a) 50 to 50% W/O emulsion (b) 20 to 80% W/O emulsion.

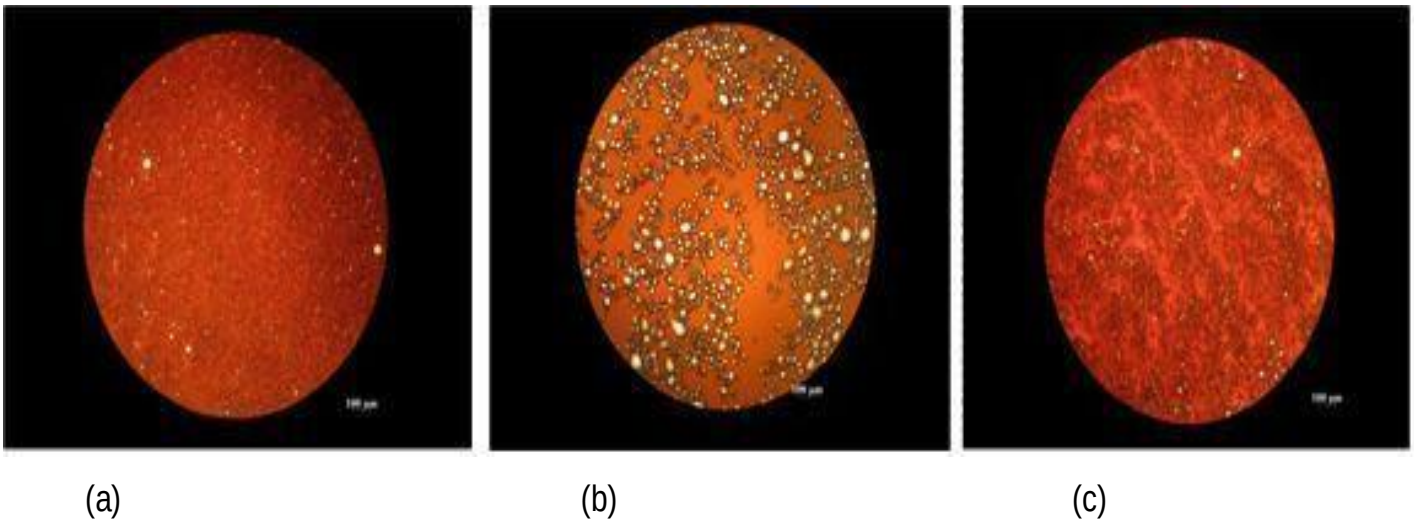

Figure 5. Microscope image of an emulsion in crude oil A, B and C before demulsified in microwave

and e) showed the droplets diameter before demulsified in microwave irradiation whereas emulsion in crude oil A and C obtained 6.59 and 5.04 μm smaller than three times emulsion prepared in crude oil B (18.59 μm). The droplet's diameter increases after demulsified in microwave irradiation as postulated by Holtze et al. (2006), the heating increases with droplets size diameter. The droplet's diameter of emulsion increases after demulsified in microwave heating which for crude oil A, the mean droplets increase up to 12.1 μm. While for crude oil B, the mean droplet's diameter showed there was a slight increase of about 19.5 μm due to the reason that the droplets were expected to coalescence each other and only small droplets are observed through microscope using 10 × resolutions. Indeed, when studying the distribution of droplets, emulsion in crude oil B obtained a uniform distribution as compared to demulsified emulsion due to the volumetric heating arising in microwave irradiation. From the figure, the droplet's diameter from crude oil C cannot be examined because emulsion was still viscous and accumulating each other thus, it was hard to measure droplet's

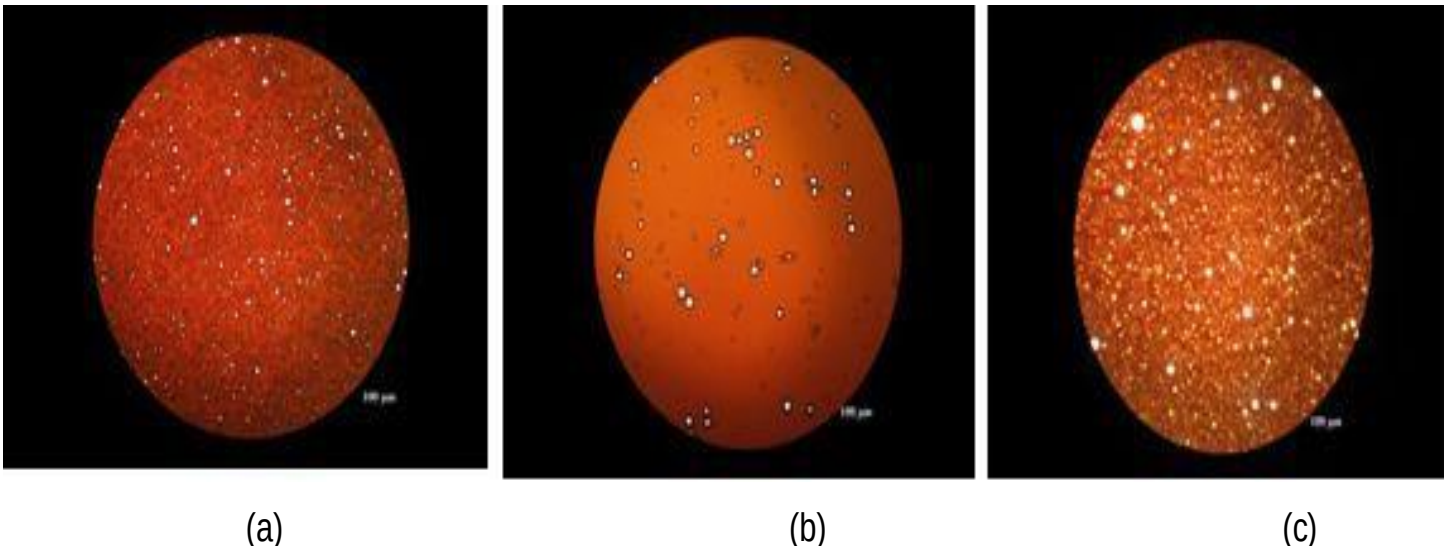

Figure 6. Microscope image of an emulsion in crude oil A, B and C after demulsified in microwave.

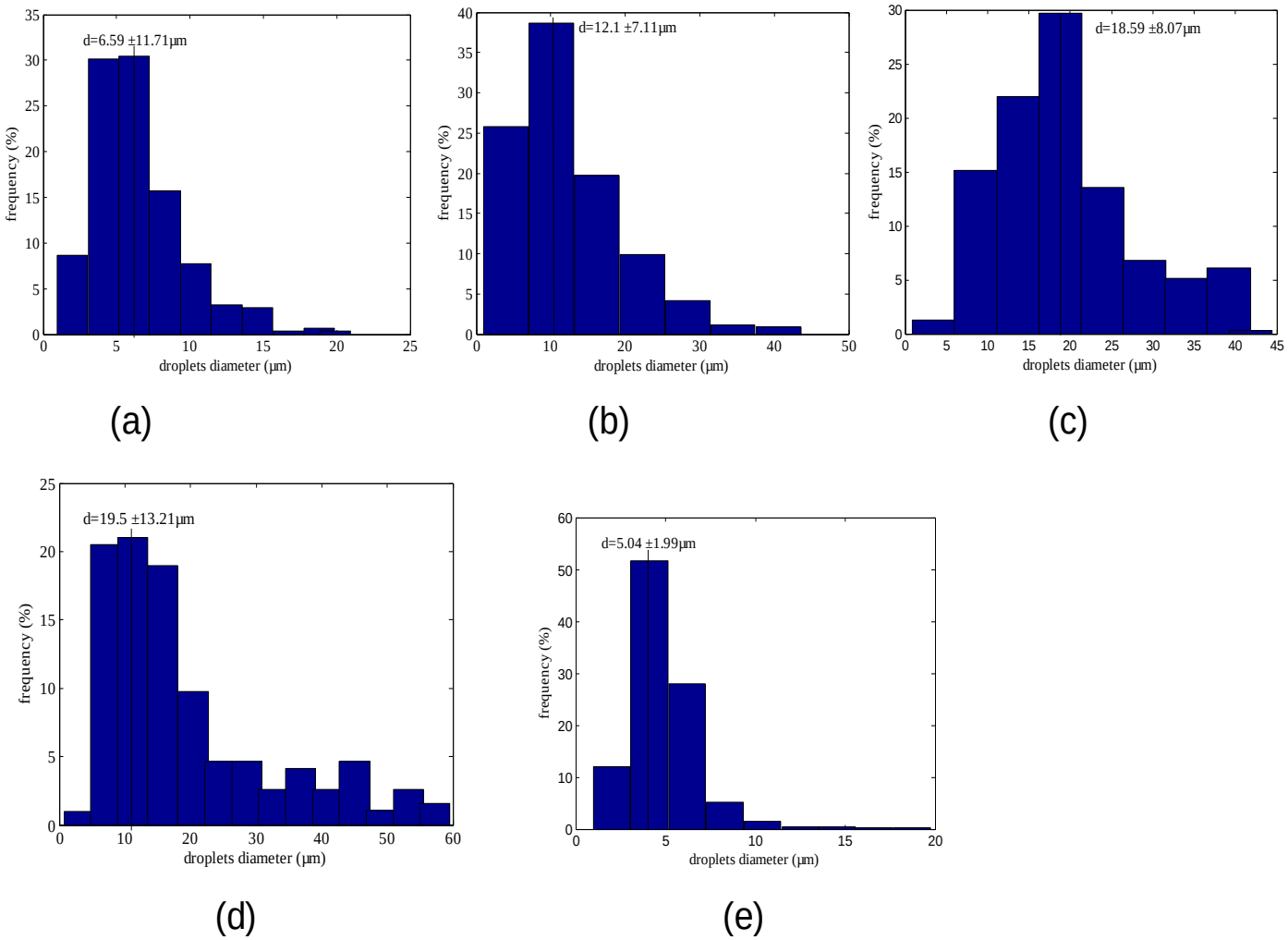

Figure 7. Droplets diameter (µm) before demulsified (a, c and e) and after demulsified (b and d) in microwave irradiation.

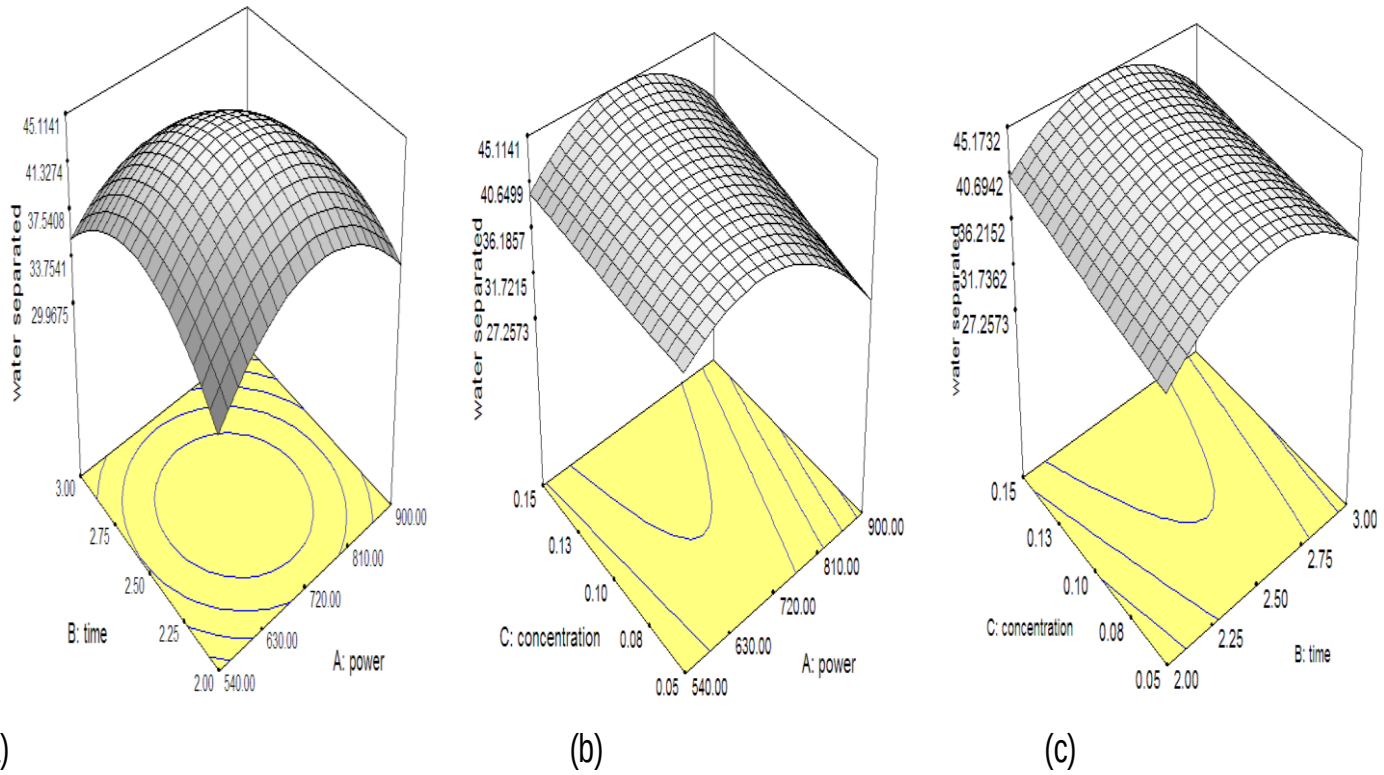

Figure 8. Response surface plotted on (a) microwave power: time processing; (b) microwave power: concentration of demulsifier; (c) time processing: concentration of demulsifier for percentage of water separated.

diameter. Besides, Urdahl et al., (1997) postulated that viscosity is higher when the size distribution is narrow due to the low degree of polydispersity.

Optimization of microwave irradiation

The performance of microwave irradiation was evaluated by observing the percentage of water separated after being demulsified in the microwave by the following equation (Jiang et al., 2007):

Water separated (%) = (Volume of water (V), ml / Original volume of water (V_o), ml) × 100 (2)

This optimization was utilizing the response surface methodology based on the central composite design of experiments as shown in Table 5. The factors were selected based on preliminary studies in screening part.

Response surface for crude oil A, B and C

Due to the preliminary experiment, crude oil A and C does not separate after demulsified in microwave irradiation because of the characteristics of the crude oil. Thus, microwave-assisted chemical system was introduced in these types of crude oil. The interactions between independent variables were plotted graphically to evaluate the percentage of water separated, based on mathematically analysis of the experimental data. The effect of microwave power, time processing and concentration of demulsifier on the percentage of water separated, as well as their interactions, are shown in Figures 8a to c. An increasing microwave power resulted in higher percentage of water separated; while the separation reached a maximum when time in microwave processing was up to a certain value, with no significant further improvement thereafter (Figures 8a and c). A different effect on the percentage of water separated was shown for concentration of demulsifier. As shown in Figures 8b and c, there was a plateau in relation to concentration of demulsifier, indicating that the percentage of water separated increased with increasing the concentration. High percentage of water separated could only be produced at higher demulsifier concentration. Molecules presented in the emulsion could be accelerated by increasing the concentration of demulsifier. Therefore, in this model, microwave power acquired high effect on the percentage of water separated because the wavelength and penetration depth increases along with microwave power. Thus, the ability of electromagnetic to penetrate into the emulsion could

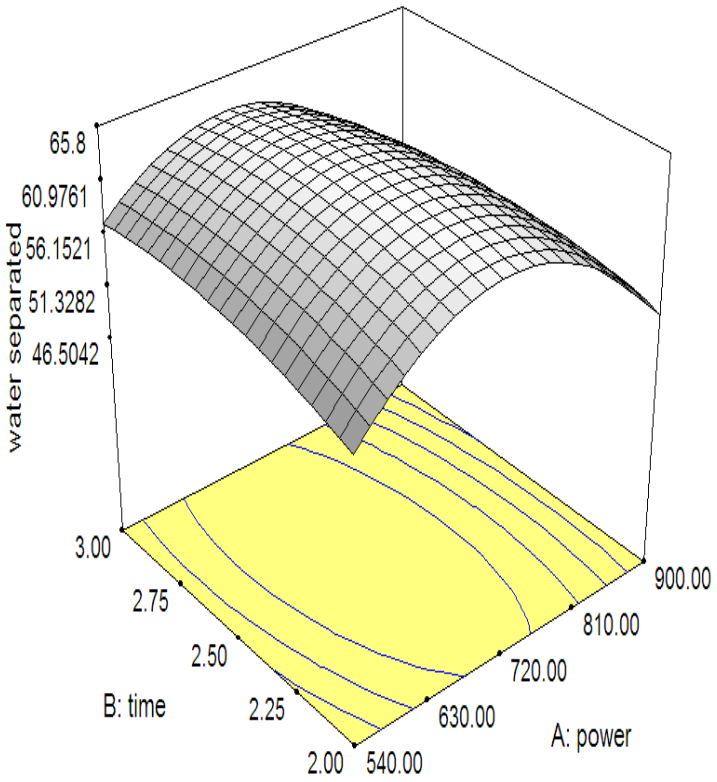

Figure 9. Response surface plots showing effects of microwave power and time processing of demulsifier on the percentage of water separation, and their interaction.

be evaluated by correlation of both penetration depth and wavelength with microwave power. Hence, a good response value, that is, 45%, was obtained at 714 W in 2.53 min and concentration of demulsifier of 0.15 wt%.

The interaction of microwave power and time processing in determining the ability of percentage of water separated in crude oil B was shown in Figure 9. From the figure, a plateau 3D graph interaction was observed in factor B (time), indicating that time do not obtain higher effect as compared to microwave power. The reason for this result was due to the characteristics of crude oil B that was acquired; a light type of crude oil. Thereby, the thermal heating occurring could be avoided when demulsified under microwave irradiation. The thermal heating was correlated to the higher temperature. In crude oil B, the light crude oil might have lower interaction of molecules as compared to heavy crude oil. Thus, at increasing time, only slight temperature increase arises in this type of crude oil. The maximum yield of water separation predicted from the response surface under microwave irradiation is illustrated in Figure 9. The response surface plot in the figure shows that this model is quadratic. The maximum yield of water separation predicted through this model is 64.0661% at microwave power 692 W in 2.56 min.

In lateral time processing, Figure 10 shows a strong response surface dependence on both microwave power and time processing. As shown in Figure 10a, there was an optimal value for microwave power and time processing in the microwave to obtain the highest percentage of water separated. Indeed, microwave power and time processing arises from a maximum point in the experimental region. However, both Figures 10b and c displayed that there was plateau in relation to the concentration of the demulsifier, which also was same in crude oil A indicating that the concentration does not affect the microwave-assisted chemical system. This is due to the higher asphaltenes presented in this crude oil. The content of asphaltenes in crude oil can be an indicator in determining the viscosity of crude oil. Crude oil C had higher viscosity increasing the demulsifier concentration in the microwave-assisted system conveyed by thermal heating in the microwave system. Thus, the concentration of demulsifier was allowed of 0.14wt% at 767 W in 2.45 min.

Analysis of variance (ANOVA) and model fitting

In general, all three factors in crude oil A and C, and two factors in crude oil B have second-order effect, that is, quadratic model on the percentage of water separated.

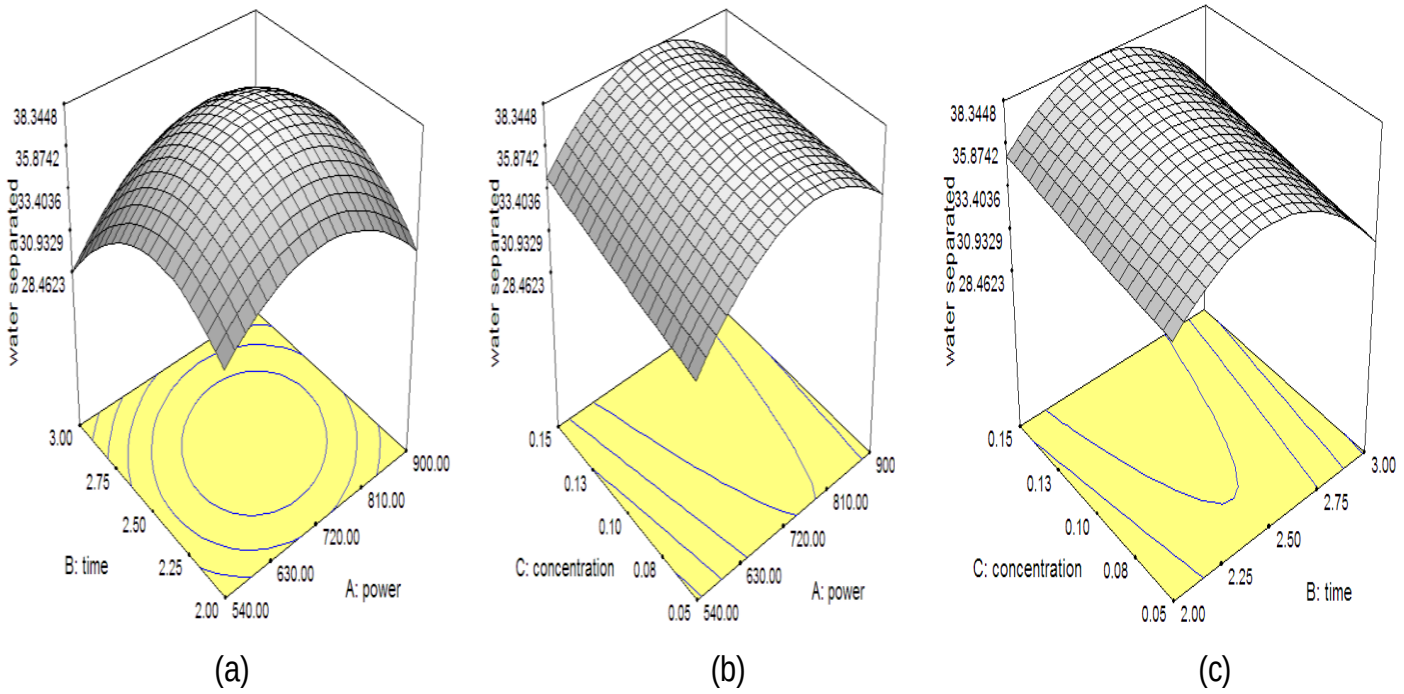

Figure 10. Response surface plotted on (a) microwave power: time processing; (b) microwave power: concentration of demulsifier; (c) time processing: concentration of demulsifier for percentage of water separated.

Table 4. Levels of the experimental variables in 2^2 factorial design.

Level of experiment variables				
Experiment variables	**α= -1**	**α= 0**	**α= +1**	**Units**
Power	540	720	900	Watt
Time	2	2.5	3	Min

The model fitted in each crude oil was analysed using analysis of variance (ANOVA) obtained from DOE. Thus, the evaluation of each crude oil was shown in Table 4, and the summary of ANOVA and regression coefficient listed in Table 5.

The quadratic models in terms of coded factors for predicting the optimal water separated are expressed as the equations 2, 3 and 4 for crude oil A, B and C in the following:

$$Y=42.9856–2.24X_1-0.34X_2+1.98\ X_3-6.1175\ X_1^2-5.4175X_2^2+0.18247X_3^2-1.7X_1\ X_2+1.35X_1\ X_3-0.6X_2X_3 \quad (2)$$

$$Y= 63.8483-2.7\ X_1+0.16667\ X_2-9.369X_1^2-1.769X_2^2-1.55\ X_1X_2 \quad (3)$$

$$Y=37.4165+1.08\ X_1-0.76X_1+0.9X_3-3.63711\ X_1^2-3.83711X_2^2+0.06289X_3^2+1.025X_1\ X_2-0.475X_1\ X_3+0.075X_2X_3 \quad (4)$$

Where Y is the percentage of water separated and X_1, X_2 and X_3 are coded variables for microwave power, time processing in microwave and concentration of demulsifier, respectively.

Results from Table 6 shows that all the three quadratic models are highly significant, implied by the high F-test values (617.457, 388.011 and 242.036) with low probability values (‘Prob>F’<0.0001). Another way to evaluate the model is lacked of fit test (Bezerra et al., 2008). A model will be well fitted to the experimental data if it presents a non-significant lack of fit. The lack of fit F-test describes the variation of the data around the fitted model (Yang et al., 2010). The lack of fit F-value from crude oil A, B and C (0.97841, 2.72748 and 2.32807) implies the lack of fit is not significant relative to the pure error. While the summary of ANOVA and regression analysis of each model listed in Table 7. Coefficient R^2 of determination is defined as the ratio of the explained variation to the total variation (Wang et al., 2008). This value indicates the relevance of the dependent variables

Table 5. Design layout and experiment response for crude oil A, B and C.

Crude oil	Std	Type	Factor A, power (Watt)	Factor B, time (min)	Factor C, concentration (wt %)	Actual, water separated (%)	Predicted, water separated (%)	Residual
	1	Fact	540	2.0	0.05	30.8	31.2830	-0.4830
	2	Fact	900	2.0	0.05	28.8	27.5030	1.2970
	3	Fact	540	3.0	0.05	34.8	35.2030	-0.4030
	4	Fact	900	3.0	0.05	24.6	24.6230	-0.0230
	5	Fact	540	2.0	0.15	34.0	33.7430	0.2570
	6	Fact	900	2.0	0.15	36.0NGN	35.3630	0.6370
	7	Fact	540	3.0	0.15	34.2	35.2630	-1.0630
	8	Fact	900	3.0	0.15	30.8	30.0830	0.7170
	9	Axial	540	2.5	0.10	40.8	39.1080	1.6920
A	10	Axial	900	2.5	0.10	32.0	34.6280	-2.6280
	11	Axial	720	2.0	0.10	36.2	37.9080	-1.7080
	12	Axial	720	3.0	0.10	38.0	37.2280	0.7720
	13	Axial	720	2.5	0.05	40.8	41.1880	-0.3880
	14	Axial	720	2.5	0.15	44.6	45.1480	-0.5480
	15	Center	720	2.5	0.10	40.2	42.9856	-2.7856
	16	Center	720	2.5	0.10	42.8	42.9856	-0.1856
	17	Center	720	2.5	0.10	45.0	42.9856	2.0144
	18	Center	720	2.5	0.10	44.6	42.9856	1.6144
	19	Center	720	2.5	0.10	44.2	42.9856	1.2144
	1	Fact	540	2.0	-	54.4	53.6937	0.7063
	2	Fact	900	2.0	-	52.6	51.3937	1.2063
	3	Fact	540	3.0	-	57.0	57.1270	-0.1270
	4	Fact	900	3.0	-	49.0	48.6270	0.3730
	5	Axial	540	2.5	-	56.6	57.1793	-0.5793
	6	Axial	900	2.5	-	50.2	51.7793	-1.5793
B	7	Axial	720	2.0	-	60.0	61.9126	-1.9126
	8	Axial	720	3.0	-	62.0	62.2460	-0.2460
	9	Center	720	2.5	-	65.8	63.8483	1.9517
	10	Center	720	2.5	-	64.2	63.8483	0.3517
	11	Center	720	2.5	-	63.6	63.8483	-0.2483
	12	Center	720	2.5	-	64.8	63.8483	0.9517
	13	Center	720	2.5	-	63.0	63.8483	-0.8483
	1	Fact	540	2.0	0.05	28.6	29.4102	-0.8102
	2	Fact	900	2.0	0.05	30.0	30.4702	-0.4702
	3	Fact	540	3.0	0.05	26.2	25.6902	0.5098
	4	Fact	900	3.0	0.05	30.2	30.8502	-0.6502
	5	Fact	540	2.0	0.15	32.4	32.0102	0.3898
	6	Fact	900	2.0	0.15	30.4	31.1702	-0.7702
C	7	Fact	540	3.0	0.15	28.8	28.5902	0.2098
	8	Fact	900	3.0	0.15	32.4	31.8502	0.5498
	9	Axial	540	2.5	0.10	32.4	32.6994	-0.2994
	10	Axial	900	2.5	0.10	36.2	34.8594	1.3406
	11	Axial	720	2.0	0.10	36.0	34.3394	1.6606
	12	Axial	720	3.0	0.10	32.2	32.8194	-0.6194
	13	Axial	720	2.5	0.05	38.0	36.5794	1.4206
	14	Axial	720	2.5	0.15	38.0	38.3794	-0.3794

Table 5. Contnd.

15	Center	720	2.5	0.10	36.8	37.4165	-0.6165
16	Center	720	2.5	0.10	36.0	37.4165	-1.4165
17	Center	720	2.5	0.10	38.0	37.4165	0.5835
18	Center	720	2.5	0.10	38.0	37.4165	0.5835
19	Center	720	2.5	0.10	36.2	37.4165	-1.2165

Table 6. Analysis of variance (ANOVA) for response surface in crude oil A, B and C.

Crude Oil	Source	Sum of squares	DF	Mean square	F value	Prob > F	Variable
A	Model	617.457	9	68.60633	18.2351	< 0.0001	significant
	A	50.176	1	50.176	13.33644	0.0053	
	B	1.156	1	1.156	0.307257	0.5929	
	C	39.204	1	39.204	10.42016	0.0104	
	A2	102.2575	1	102.2575	27.17934	0.0006	
	B2	80.19464	1	80.19464	21.31519	0.0013	
	C2	0.09098	1	0.09098	0.024182	0.8799	
	AB	23.12	1	23.12	6.14514	0.0351	
	AC	14.58	1	14.58	3.875265	0.0805	
	BC	2.88	1	2.88	0.765485	0.4044	
	Residual	33.8609	9	3.76232			
	Lack of Fit	18.6289	5	77.6023	0.97841		not significant
B	Model	388.011	5	43.74	38.0462	<0.0001	significant
	A	43.74	1	0.16667	21.4445	0.0024	
	B	0.16667	1	242.433	0.08171	0.7833	
	A2	242.433	1	8.64266	118.858	<0.0001	
	B2	8.64266	1	9.61	4.23725	0.0785	
	AB	9.61	1	2.03969	4.7115	0.0666	
	Residual	14.2778	7	3.19661			
	Lack of Fit	9.58982	3	1.172	2.72748	0.1784	not significant
C	Model	242.036	9	26.8929	16.8208	0.0001	significant
	A	11.664	1	11.664	7.29551	0.0244	
	B	5.776	1	5.776	3.61273	0.0898	
	C	8.1	1	8.1	5.06633	0.0509	
	A2	36.1457	1	36.1457	22.6082	0.0010	
	B2	40.2302	1	40.2302	25.1629	0.0007	
	C2	0.01081	1	0.01081	0.00676	0.9363	
	AB	8.405	1	8.405	5.2571	0.0476	
	AC	1.805	1	1.805	1.12898	0.3157	
	BC	0.045	1	0.045	0.02815	0.8705	
	Residual	14.3891	9	1.59879			
	Lack of Fit	10.7091	5	2.14182	2.32807	0.2166	not significant

in the model which the small value of R^2 shows that poor relevance of the dependent variables. From this study, the correlation coefficient (R^2) values of three models are 0.9480, 0.9645 and 0.9439 which the second-order model explained about 94.80, 96.45 and 96.39% of the variability observed in the gain, indicating a

Table 7. Summary of ANOVA and regression analysis for each crude oil.

Crude oil	Significant model of terms	Standard deviation	R^2	Adjusted R^2	Predicted R^2	Adequate precision
A	A, C, A2, B2, AB	1.9397	0.9480	0.8960	0.7330	14.5859
B	A, A2	1.4282	0.9645	0.9392	0.7968	15.6879
C	A, A2, B2, AB	1.2644	0.9439	0.8878	0.6229	13.8330

satisfactory fitting of the quadratic models to experimental data. The adjusted R^2 for model A, B and C are 0.8960, 0.9392 and 0.8878, which considered as good fit for the observed response values. Values Prob >F less than 0.05 indicated model terms are significant. In this case, A, C, A2, B2 and AB are significant model terms. While for crude oil B, factor A and A2 is significantly model terms and in crude oil C, factor A, A2, B2 and AB display the significant model terms.

Indeed, an adequate precision compares the range of the predicted values at the design points to the average prediction error. In this study, for all three models, the values of an adequate model were 14.5859, 15.6879 and 13.8330; which greater than 4, indicating adequate model's discrimination.

Conclusions

Comparing conventional heating and microwave irradiation, the demulsification process efficiency increased in the microwave due to the viscosity reduction. Besides, the droplets size distribution in crude oil B also supported that microwave irradiation enhanced the demulsification rather than using conventional heating. In optimization part, the models exhibited a quadratic polynomial model could be employed to optimize the water separated in microwave irradiation. From response surface graph, microwave power and time significantly influenced the percentage of water separated in heavy crude oil (crude oil A and C). While for crude oil B, the processing time in the microwave does not reveal as high as microwave power effect in percentage of water separated.

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