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Elucidate the Separation Mechanisms of Continuous Microwave Radiation Systems (CMWRS) Using Crude Oil Emulsions

N.H.Abdurahman*1, N.H.Azharib2, Y.M.Roslic3
[*1,3] Faculty of Chemical and Natural Resources Engineering, University Malaysia Pahang
[2] Faculty of Industrial Sciences and Technology, University Malaysia Pahang
nour2000_99@yahoo.com

Abstract

The crude oil is an indispensable source of energy. During the transportation, lifting and processing of oil, water-oil emulsions are frequently created. The traditional methods of eliminating these emulsions are based on utilization of high heat, chemicals, and electrical energy to force the emulsion to separate into water and hydrocarbon phases are disadvantages from both economic and environmental perspectives. In this work, a continuous microwave radiation system was developed and applied successfully for separation of heavy crude oil emulsions.

In this study, a continuous microwave demulsification method was utilized in a 50-50%, and 20-80% of water-in-oil emulsions with varied microwave exposure time. Temperature profiles of water-in-oil emulsions inside a cylindrical container were measured. The temperature rise at a given location was linear. The rate of temperature increase of emulsions decreased at higher temperature due to decreasing dielectric loss of water. Due to its fast, volumetric, and selective heating, microwave heating can be used an alternative demulsification method for water-in-oil emulsions.

Keywords: Continuous microwave, separation, w/o emulsions, radiation, stability, elucidate

Introduction

Microwaves are electromagnetic waves with a frequency of 300 MHz to 300 GHz, corresponding to wavelengths of 1 mm to 1 m. Beyond 30 GHz; the microwave frequency range overlaps with the radio frequency. Because of the nature of microwaves, they are governed by the Federal Communications Commission (FCC). In North America, the only allowed frequencies for industrial use are 915, 2450, 5800 and 22,000 MHz (Dumbaugh et al, 2001). For use in Laboratory reactions, a frequency of 2450 MHz is preferred, since this frequency has adequate penetration depth for most laboratory reaction conditions.

Microwave energy is receiving a considerable amount of attention from researchers for a wide spectrum of applications (Belanger et al., 2008; Whittaker, 2009; Will et al., 2004). The fundamentally different method of transferring energy from the source to the sample is the main benefit of utilizing microwave energy when compared to conventional thermal processing. The crude oil containing water is harmful to the transportation, refinery, and quality of the products so that demulsification is one of the key steps in various sectors of the oil industry, including crude oil transportation, petroleum refinery, as well as oil production. Two principal approaches of demulsification are chemical and physical methods. The chemical method is the addition of proper demulsifier to the emulsion and typical physical treatment techniques include heating, electrical, ultrasonic, or mechanical method such as centrifugation. The heating along with the addition of the demulsifier (thermal chemical method) and electrical techniques are most popular methods, but these methods have many disadvantages: power-wasting, a large addition of the demulsifier, bad water quality, and environmental pollution.

Microwave has drawn much more attention as a clean and efficient energy in recent years; many papers reported by (Fang, et al., 1995, 1988; Xia et al., 2003; Liu et al., 2005; Chih et al., 2002; Vladana et al., 2006) have reported the advantages of this new technology after it was first tested by Klaila (1978) and Wolf (1986). However, the reports about the practical application of microwave demulsification are very limited up to now.

Materials and Methods

In this study, Elba domestic microwave oven model: EMO 808SS, its rated power output is 900 watts and its operation frequency is 2450 MHz was modified and converted from batch process system to a continuous process and used as shown in Figures 1 and 2 respectively.

Figure 1- Continuous microwave process

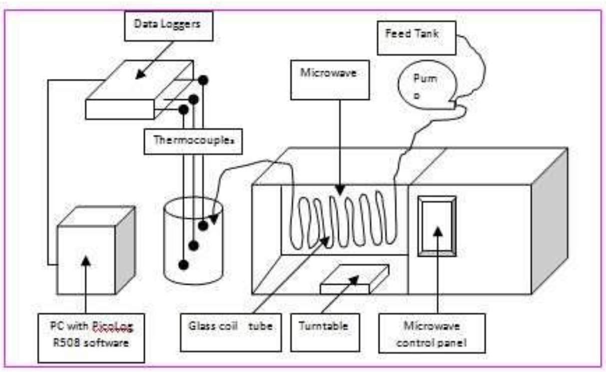

Figure 2- Continuous microwave processes

Three thermocouples type (K-IEC-584-3) were connected to Pico-TC-08 data loggers, and then the thermocouples connected to the settling tank. The data logger was connected to Pc; with Pico Log Rs.08.3 software. The thermocouples were inserted in the settling tank to different locations top, middle, and bottom of the emulsion sample to measure local temperatures.

Sample Preparation and Procedures

The crude oil samples were obtained from Petronas refinery at Melaka city, 50-50%, and 20-80% water-in-oil emulsions were prepared. Emulsions were prepared in 900 ml graduated beakers, with ranges by volume of the water and oil phase. The microwave radiation was set to different power settings. The water phase is distilled water. The emulsions were agitated vigorously using a standard three blade propeller at speed of 2000 rpm and temperature 30 ^{o}C for 15 minutes. The concentrations of water in samples were 20-50% by volume. The volume of water settled to the bottom was read from the scale on the beaker with different times. The prepared emulsions were used to check for w/o or o/w emulsions. All emulsions investigated were type of water-in-oil emulsions (oil continuous phase). Surfactant used in this study is sodium carbonate (Na_2CO_3).The sample placed in a feed tank and used a pump to pull the samples to the Elba domestic microwave oven model: EMO 808SS for radiation. Three thermocouples were inserted in the settling tank of emulsion sample at different locations, top, middle, and bottom. The emulsion samples were heated with microwave radiation for 20, 40, 60, 80, 100, 120, 140, 160, 180, and 200 seconds. Temperature profiles of emulsions inside a cylindrical container during continuous microwave heating at 2450 MHz were recorded by Pico-TC-08 data logging.

Microwave Radiation

A number of studies were carried out on microwave heating (MW) of oil and water systems. Microwave heating because of its volumetric heating effects, offers a faster processing rate, also microwave has another unique feature other than how it interacts with matter, is its ‘’penetrating power’’. Microwave distributes energy within the bulk of most materials, rather than just on it surface. Any heat produced at the surface must then be conducted or convicted into the material. Microwave, because the wave length is relatively long and the method of interaction so mild, can penetrate deeply into a substance. Penetration energy deposition by microwave overcomes many surface-limiting characteristics of normal heating. The purpose of heating water-in-oil (w/o) emulsions with microwave radiation is to separate water from oil. Therefore, when water-in-oil emulsion is heated with microwave radiation, two phenomenons will occur; the first one is the increase of temperature, which causes reduction of viscosity and coalescence. The result is separation of water without addition of chemicals (Fang, at al., 1988, 1989). According to Stoke’s law Eq (1), if oil is the continuous phase, the settling velocity of water droplets is given by:

$$v_w = \frac{(\rho_w - \rho_o) g D^2}{18 \mu_o} \qquad (1)$$

where D is the diameter of the droplets. The viscosity of oil very sensitive to temperature, as temperature increases, viscosity decreases much faster than the density difference, $(\rho_w - \rho_o)$ does, the result when viscosity decreases, the droplets size increases. Therefore, microwave heating increases the velocity of water (v_w), and accelerates the separation of emulsion. The higher temperature and lower viscosity make the coagulation process easier. The results are larger particle diameter D, and rapid separation. Since microwave heats materials volumetrically, it is possible to calculate the volume rate of microwave heat generation from energy balance equation (2) as:

$$g_{MW}=\frac{hA}{V}(T_m-T_a)+\frac{\varepsilon A\sigma}{V}\left[(T_m+27315)^4-(T_a+27315)^4\right]+\rho C_p\left(\frac{dT}{dt}\right) \quad (2)$$

The above equation assumes that the rate of heat transfer from emulsified water droplets to the continuous phase (oil) is very rapid; therefore, water and oil practically have the same temperature. The right hand side of Equation (2) comprises of three terms, convective heat transfer, irradiative heat due to microwave, and conductive heat in the sample respectively. From results of this study, the effect of radiative term is very small as well as convective term. Since the sample container (glass) has low dielectric constant, therefore, its heat generated assumed to be negligible. For calculation of volume rate of heat generation in Equation (2), the density (ρ), and (C_p) of the emulsions calculated from mixing rules Eqs. (3) & (4) as:

$$\rho_m = \rho_w\phi + \rho_o(1-\phi) \quad (3)$$

$$C_{pm} = C_{p,w}\phi + C_{p,o}(1-\phi) \quad (4)$$

The volume rates of microwave heat generation of the 50-50% and 20-80% water-in-oil emulsions calculated from temperature measurements and Equation (2) were shown in Tables 1 & 2 respectively. While viscosity data were shown in Table 3.

Table1. Experimental results of continuous microwave heating (50-50% w/o emulsions) (Microwave power is: 900W)

Radiation time, sec	T_o=25.6 C Δ T, °C	Rate of Temp. increase dT/dt, C/sec	Rate of Heat generation q_{MW} $\frac{cal}{\sec - cm^3}$	Dielectric constant $\varepsilon'_{r,50-50\%}$	Dielectric loss $\varepsilon''_{r,50-50\%}$	$\tan\delta = \frac{\varepsilon''_r}{\varepsilon'_r}$
20	15	0.75	0.494	39.70	11.359	0.286
40	18	0.45	0.297	39.198	8.245	0.210
60	21.5	0.36	0.237	38.610	6.870	0.178
80	26	0.33	0.217	37.852	5.653	0.149
100	30.5	0.31	0.204	37.095	4.799	0.129
120	33.7	0.28	0.185	36.557	4.332	0.118
140	37.5	0.27	0.178	35.917	3.882	0.108
160	41.0	0.26	0.171	35.328	3.543	0.100
180	44.6	0.25	0.165	34.722	3.250	0.094
200	47.8	0.24	0.158	34.184	3.027	0.089

Table2. Experimental results of continuous microwave heating (20-80% w/o emulsions) (Microwave power is: 900W)

Radiation time, sec	T_o=25.6 C Δ T, °C	Rate of Temp. increase dT/dt, C/sec	Rate of Heat generation q_{MW} $\frac{cal}{\sec - cm^3}$	Dielectric constant $\varepsilon'_{r,20-80\%}$	Dielectric loss $\varepsilon''_{r,20-80\%}$	$\tan\delta = \frac{\varepsilon''_r}{\varepsilon'_r}$
20	19	0.95	0.474	16.948	3.122	0.184
40	24	0.60	0.299	16.609	2.457	0.148
60	27	0.45	0.225	16.406	2.178	0.133
80	34	0.43	0.215	15.932	1.720	0.108
100	40	0.4	0.200	15.525	1.457	0.094
120	45	0.38	0.190	15.186	1.292	0.085
140	51	0.36	0.180	14.780	1.137	0.077

160	57	0.36	0.180	14.373	1.016	0.070
180	62	0.34	0.170	14.035	0.933	0.066
200	67	0.34	0.170	13.696	0.862	0.063

Dielectric constant and dielectric loss of water used in this work were given by [Von Hippel, 1954] as follows:

$$\varepsilon'_r = 85.215 - 0.33583T \quad (5)$$

$$\varepsilon''_r = 320.658T^{-1.0268} \quad (6)$$

Von Hippel (1954) proposed equations for dielectric properties of various petroleum oils, in this regards, dielectric constant and loss tangent of crude oil for this study calculated from the equations below.

$$\varepsilon'_{r_o} = 2.24 - 0.000727\,T \quad (7)$$

$$\tan\delta_o = (0.527T + 4.82) * 10^{-4} \quad (8)$$

Table 3 shows viscosity data for 50-50% and 20-80% water-in-oil emulsions respectively.

Table 3: Viscosity data for 50-50% and 20-80% w/o emulsions

Viscosity (Pa.s)	Temperature © °C	Shear rate (sec^{-1})	Shear stress ($dyne/cm^2$)
50-50 % w/o 0.490	25	35	29
0.350	40	40	32.8
0.300	50	45	35
0.260	60	51	38
0.200	70	58	43
0.175	80	65	48
20-80 % w/o 0.209	25	32	23.5
0.190	40	39	24.6
0.177	50	37	26.5
0.120	60	41	30.9
0.089	70	47	37.5
0.070	80	50	40

Results and Discussion

The microwave heating process was examined for 50-50% and 20-80% of crude oil emulsion samples. Transient temperature profiles of water-in-oil emulsions during a continuous microwave heating were measured. The results of coalescence between liquid droplets of 50-50% and 20-80% water-in-oil emulsions were shown in Figures 3 and 4 respectively. All experimental results showed that microwave radiation is very effective in separation of water-in-oil emulsions. Figures 3 and 4 illustrated that, microwave radiation can raise the temperature of emulsion, reduce the viscosity, increase the velocity and accelerate separation process as suggested by Equation (1). It is found that the sodium carbonate (Na_2CO_3) was very efficient in stabilizing water-in-oil emulsions. The percent of coalesced or separated water is plotted against the time of sedimentation. It observed that, the 50-50% w/o was separated faster than 20-80% does, this may attributed due to high volume fraction for 50-50% ($\phi = 0.46$) compared to 20-80%. ($\phi = 0.18$). It found that the percentage of coalesced water droplets decreases with the concentration of the sodium carbonate (Na_2CO_3).

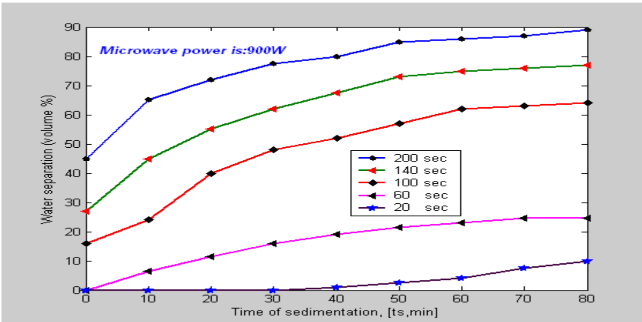

Figure 3: Separation of water from 50-50% water-in-oil emulsion

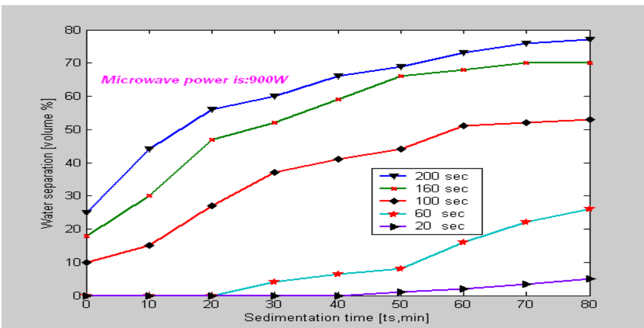

Figure 4: Separation of water from 20-80% water-in-oil emulsion emulsions

The temperature increasing rates of irradiated samples, dielectric constant, dielectric loss, loss tangents, and volume rates of heat generation for 50-50% and 20-80% w/o emulsions were shown in Tables 1 and 2 respectively. The rate of temperature increase was calculated from temperature increase divided by radiation time. It is observed that, the rate of temperature increase $(\frac{dT}{dt})$ is inversely proportional to the increase in temperature ΔT; this was expected result since the dielectric loss of water is small. The rates of temperature increase for 50-50% and 20-80% w/o illustrated in Figure 5. Equation (2) used to calculate the volume rate of heat generation, from the calculations; it found that the contributions of the heat loss by convective heat transfer and radiative heat loss were very small, while the contribution of heat accumulation in the emulsion is significant.

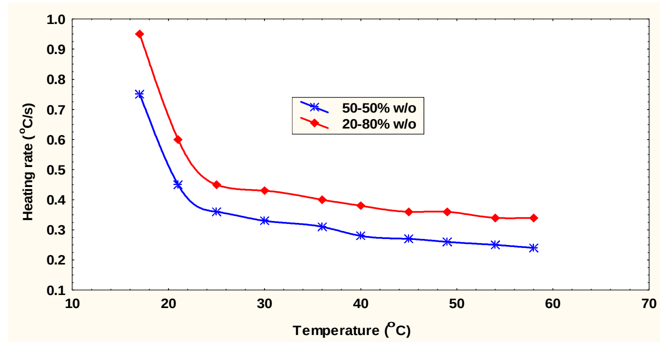

Figure 5: Rates of temperature increase for 50-50% and 20-80% w/o emulsions

In application of Equation (2) for calculation of volume rates of heat generation, the emulsion density (ρ_m) and heat capacity (cp_m) were calculated from mixture rules Equations (3) and (4) respectively. It is observed that, the dielectric properties of emulsions affected by temperature in this regards, Figure 6 shows dielectric losses for 50-50% and 20-80% w/o emulsions. It is clear from the figure, dielectric loss for 20-80% w/o less than for 50-50% this may attributed to the high temperature of 20-80% compared with temperature of 50-50% w/o.

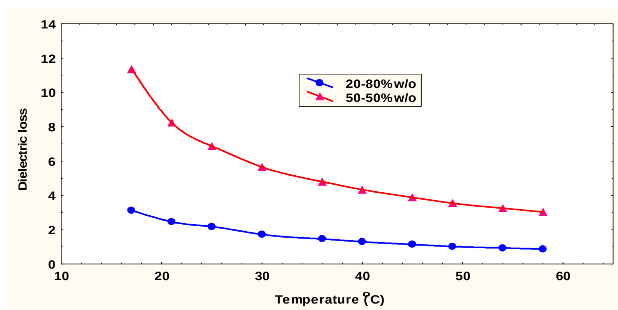

Figure 6: Dielectric loss vs. Temperatures for 50-50% and 20-80% w/o emulsions

The shear rate, shear stress, and viscosity of the emulsion samples were measured with Brookfield (DV-III) Rheometers given in Table3. The viscosity, μ of an emulsion diminishes when the volume fraction of the dispersed phase ϕ is reduced.

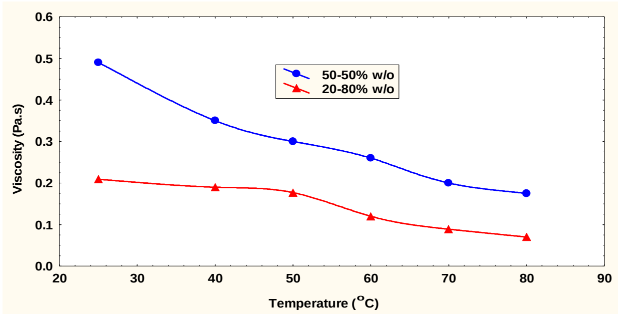

Figure7: Viscosity vs. temperature for 50-50% and 20-80% w/o emulsions

Figure 7 shows the viscosity of w/o versus temperature, it's clear from the Figure 7; emulsions were very sensitive to temperature. As temperature increases, the viscosity decreases fast. The viscosities for 50-50% and 20-80% for w/o emulsions are shown in Figure 8. Increases in the internal phase volume fraction lead to an increase in both the viscosity and the degree of shear thinning.

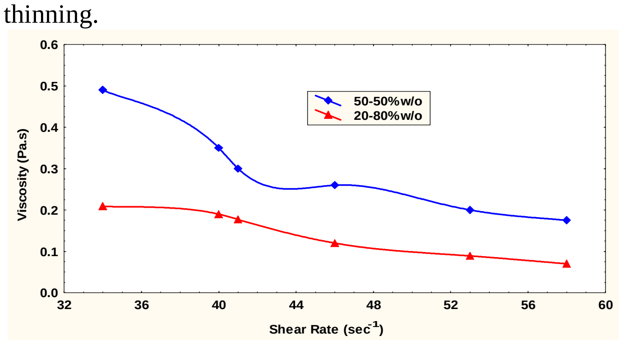

Figure 8: Viscosity vs. Shear rate for 50-50% and 20-80% w/o emulsions

Conclusions

A continuous microwave heating system was developed and successfully examined on crude oil emulsions. This study has shown that microwave radiation can be an effective tool to separate water from dispersed water-in-oil emulsions. Microwave heating provides a new option in breaking water-in-oil emulsions and enhances gravity sedimentation to separate the emulsions into water and oil layers.

Microwave separation technology does not require chemical addition. Consequently, it is an attractive alternative to the conventional method which sometimes requires a heavy dosage of chemicals. The temperature rise and volume rate of heat generation of emulsions induced by microwave radiation can be calculated from basic dielectric properties.

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