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Correlations between interfacial tension and cumulative tertiary oil recovery in a triglyceride microemulsion flooding

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ABSTRACT

This paper presents measurements of interfacial tension (IFT) and tertiary oil recovery (TOR) of triglyceride microemulsion flooding over a wide range of aqueous phase compositions. Based on 160 experimental data sets, two empirical correlations were established. Both the power-law and logarithmic models were validated statistically. Power-law and logarithmic models are predicted to perform best at ultralow IFT range (<0.001 mN/m) and high IFT range (>2 mN/m), respectively. The valid models indicate that IFT is the sole parameter affecting the cumulative TOR in a triglyceride microemulsion flooding. This phenomenon, however, does not apply in hydrocarbon-based microemulsions.

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1. Introduction

Oil, water and an amphiphile mixture are the main components of a microemulsion [1]. Microemulsion flooding, which is a promising technique of tertiary oil recovery (TOR), is conducted in laboratory by injecting slug of microemulsion slug post water flooding into a porous medium. The porous medium can either be a sand pack or core. Over the past several decades, numerous successful applications of various microemulsion formulations in enhanced oil recovery (EOR) have been reported [2–5].

The success of a microemulsion flooding relies highly on the interactions between the displacing fluid (microemulsion) and remaining oil [6]. Thus the efficiency of a microemulsion formulation depends on the amount and chemical nature of its components [6,7]. A change in the composition or chemical nature of microemulsion leads to an alteration of fluid–fluid interactions at the interface or the interfacial tension (IFT) between the displacing fluid and the remaining oil [6,8]. IFT variation subsequently affects the type of in situ microemulsion and amount of recovered oil [8–11]. It is believed that the displacement efficiency increases remarkably with IFT reduction [9]. The larger oil recoveries were observed when a microemulsion slug produces lower IFT against the remaining oil [9]. Attempts have been made

recently to formulate new microemulsion with ultralow IFT to improve oil recovery [7,12,13]. Therefore, it is evident that IFT plays a critical role on the amount of the oil recovery.

The effect of numerous parameters and variables on IFT and consequently on oil displacement efficiency has been investigated experimentally for several decades [9,14–19]. Based on these studies, many new empirical correlations were developed [16,17,19]. Based on these studies it was concluded that ultralow IFT is not the sole condition required ensuring improvement of oil production in TOR [17]. Beside IFT, interfacial viscosity and the dynamic process of surfactant partitioning are two other parameters which have critical role in mobilization of the trapped oil after the secondary recovery [17]. In addition, the capillary number, which is expressed as the ratio of viscous to capillary forces, was introduced as a correlation that governs the immiscible displacement and mobilization of the remaining oil from rock samples and sand packs [20–23].

The relationship of IFT and oil recovery efficiency is wellestablished in the literature for common hydrocarbon-based microemulsion systems. However, works related to triglyceride microemulsion are very scarce. A triglyceride microemulsion has been formulated by having the triglycerides for the whole oilphase of a microemulsion [10,24–32]. The potential of the application of triglyceride microemulsion in EOR has recently been studied [5,7,33]. It is believed that the change of the oil component of a microemulsion from hydrocarbon fractions to triglyceride affects the parameters, which lead to high TOR. Therefore, an empirical correlation of the IFT and cumulative TOR

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could provide information on the performance of a triglyceride microemulsion flooding.

The objective of this paper is to investigate the effect of IFT on oil recovery using a triglyceride microemulsion formulation. In the triglyceride microemulsion formulation, palm oil was used as the oil phase of the microemulsion. Alkyl polyglycosides (APG) and glyceryl monooleate (GM) were used as the surfactant and cosurfactant of the triglyceride microemulsion, respectively.

2. Experimental tests

2.1. Materials

The surfactant used in this work was Glucopon 650EC, which is a mixture of alkyl polyglycosides (APGs), having an average alkyl chain length of 11, hydrophilic–lipophilic balance (HLB) of 11.9, and critical micelle concentration (CMC) of 0.073 g/L at 37 °C [34]. It was supplied by Cognis (Malaysia) Sdn Bhd which is part of BASF Chemical Company. The active percentage of the surfactant solution is 50–53 weight percent (wt%).

Sodium chloride (NaCl, A.R. grade) was supplied by LGC Scientific, Malaysia. N-octane (free of olefins) and glyceryl monooleate (GM) were supplied by Sigma–Aldrich. Palm kernel oil was purchased from Delima Oil Products Sdn. Bhd. in Malaysia. The palm kernel oil contains 43.33 wt% monounsaturated fat, 12.22 wt% polyunsaturated fat, and 44.45 wt% saturated fat. Compared to other vegetable oils, palm oil is more abundant and widely available in Southeast Asia, particularly in Malaysia. In comparison with other palm oils, palm kernel oil is much cheaper and it is preferred for industrial and non-food applications. All of the materials were used as supplied with no additional purification.

2.2. Triglyceride microemulsion preparation

The oil phase of the triglyceride microemulsion samples was pure palm oil. The aqueous phase of the triglyceride microemulsion samples was composed of APG, GM, NaCl, and de-ionized water at various compositions. The concentrations of APG in the aqueous phase were 0.5, 1, 1.5, and 2 wt%. The concentrations of NaCl in the aqueous phase were 1, 3, 6, 9, and 12 wt%. The concentrations of GM in the aqueous phase were 0.5, 1, 1.5, 2, 2.5, 3, 3.5, and 4 wt%. Thus 160 Winsor Type I triglyceride microemulsion samples were prepared and separated from the upper (excess oil) phase with the same procedure as in our previous works [7,33]. Then they were used in the next measurements.

2.3. IFT measurements

IFT between the heavy and light phases were measured and recorded at constant temperature of 30 °C. In all of the IFT measurements, the heavy phase was the triglyceride microemulsion phase previously prepared. The light phase was n-octane, which represents the crude oil in reservoirs [35]. IFTs were measured using a Spinning Drop Tensiometer Site 4 (KRUSS, Germany) based on the same procedure described in our previous works [7,36].

2.4. Microemulsion flooding

Microemulsion flooding tests were conducted to determine the performance of all of the formulated microemulsions in TOR. The same sand and sand pack were used in these experiments as in our previous work [7]. The same flooding procedure was also used until the residual oil saturation after the secondary oil recovery by water flooding was reached. In the tertiary recovery, the sand pack was continuously flooded with 4 PV of a prepared triglyceride microemulsion. The produced oil in TOR was recorded by GC as explained elsewhere [7].

2.5. Application of MATLAB in curve fitting

MATLAB software provides an advanced curve fitting toolbox to facilitate both parametric and nonparametric data fitting. A parametric fit can be performed using either a toolbox library equation or a custom equation. Library equations include polynomials, exponentials, rationals, sum of Gaussians, etc. In addition, a custom equation can be defined by user based on the specific needs of fitting.

In this paper, MATLAB software version 7.2.0.232 (MathWorks, Inc., Natick, Massachusetts, U.S.A.) was used to conduct statistical parametric fitting. All library equations and many custom equations were tested to capture the relationship between 160 experimental data sets of IFT and cumulative TOR. The goodness of fit was evaluated by the fit statistics provided in the software.

3. Results and discussion

3.1. Experimental results

Table 1 shows all of the 160 experimental data sets of IFT and cumulative TOR as a function of the aqueous phase composition of the microemulsion. These data were obtained by IFT measurements and microemulsion flooding.

3.2. Modeling using MATLAB

To investigate the effect of IFT on cumulative TOR, all of the 160 experimental data sets of IFT and cumulative TOR were used regardless of the aqueous phase composition. First, all the equations available in the library of MATLAB were tested for fitting. Among all the equations, only power-law model is able to capture a relationship between IFT and cumulative TOR. The typical power-law model is expressed as follows:

$$y = a x^b \tag{1}$$

Considering the *y*-data for cumulative TOR and *x*-data for IFT, the coefficients of the power-law model of cumulative TOR as a function of IFT and the statistical parameters showing the goodness of fitting are tabulated in Table 2. The coefficients of the model were obtained by curve-fitting and nonlinear regression techniques.

The accuracy of the model can be checked statistically from the regression line between the experimental and predicted cumulative TOR by the model. Fig. 1 shows the regression line for the power-law model. Four statistical parameters of the regression line which are the sum of squares due to error (SSE), *R*-square, adjusted *R*-square, and root mean squared error (RMSE) are given in Table 2. These statistical parameters confirm the accuracy of the correlation. On the one hand, both *R*-square and adjusted *R*-square are found to be 0.9994. These values are close to one, which indicates the fitting is satisfactory. In addition, the calculated SSE and RMSE are 0.3534 and 0.04729, respectively. The small values of these parameters indicate a successful fitting.

Beside trial of all the equations in the MATLAB library, a custom equation was found to be capable of establishing adequate correlation between the experimental data sets of IFT and cumulative TOR. The custom equation, which was found by trial and error, is a modified logarithmic function stated below:

$$y = a - \log\left(\left(\frac{x}{0.0002}\right)^b\right) \tag{2}$$

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