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Characterization of water-in-crude-oil emulsions in a complex shear field

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ABSTRACT

Understanding the formation and stability characteristics of concentrated emulsions generated in complex flow fields is of importance to the energy industry. Knowledge about the transient stability of water-in-crude-oil emulsions aids in formulating flow assurance strategies to problems such as water separation and gas hydrate formation. During the production of crude oil, the produced fluids can experience a wide range of flow conditions, such as flow through pumps, reductions and expansions, and perforations which affect both the formation and stability of the emulsions. This work focuses on complex water-in-crude-oil emulsion formation and stability. The emulsions are formed with a rotating turbine using synthetic seawater as the dispersed phase and two crude oils as the continuous phase. This work employs the Carr-Purcell-Meiboom-Gill (CPMG) nuclear magnetic resonance (NMR) technique to measure the transient drop size distributions. The effects of parameters such as the mixing Reynolds number, energy input, and mixing time on emulsion formation and stability were quantified. The effect of mild shear after emulsification was also evaluated.

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1. Introduction

Formation and stability characteristics of emulsions created in complex flow fields are particularly applicable to the energy industry [1]. The transient stability of water-in-crude-oil emulsions affects both water separation and methane hydrate formation in flowing systems. The shear history of an emulsion has a profound impact on the overall behavior of the emulsion throughout its life in a process system. In addition, the presence of contaminants and natural surfactants in crude oil systems affects the stability of crude oil emulsions. Fundamental understanding and quantification of concentrated emulsion formation and stability mechanisms in complex crude oil systems is still lacking.

In the laboratory, emulsions are generated using a variety of techniques, one of which is the use of a turbine mixer such as a Rushton turbine [2]. Rushton turbines have been widely used in industry and academia to investigate mixing phenomena and drop breakup [2,3]. Rushton turbines facilitate adequate mixing of the immiscible fluids and provide sufficient shear to disperse water drops in oil. In addition, published correlations facilitate the calculation of power and energy applied to the emulsion systems using Rushton turbine mixers.

* Corresponding author. Tel.: +1 405 744 9110. E-mail address: clint.aichele@okstate.edu (C.P. Aichele). Hinze pioneered drop breakup in turbulent flow by showing that the maximum equilibrium drop size (d_{max}) was a function of the energy dissipation rate [4].

$$d_{\max} = C \varepsilon^{-0.4} \left(\frac{\sigma}{\rho_{CP}} \right)^{0.6} \tag{1}$$

In this expression, *C* is a correlation constant, ε is the energy dissipation rate, σ is the interfacial tension, and ρ_{CP} is the density of the continuous phase. Work by Hinze showed that drop breakup in a stirred tank environment primarily occurs in the impeller region due to turbulent pressure fluctuations along the drop's surface [4]. Subsequently, Chen and Middleman used similar arguments to empirically derive an expression for the Sauter diameter for dilute emulsions with an inviscid dispersed phase produced by Rushton turbines [5].

$$\frac{d_{32}}{L} = 0.053 W e^{-3/5} \tag{2}$$

The Sauter mean diameter, d_{32} , is the size of a drop which has the same volume-to-surface area ratio as the entire population of drops [6]. The impeller diameter is represented by the symbol *L* in Eq. (2). The Sauter diameter gives important information about the available interfacial area of the drops in the distribution. The Sauter diameter is commonly used in the literature to describe





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Nomenciature

a, b, C d:	empirical constants diameter of the <i>i</i> th hin (um)	$T_{2,DP,i}$	transverse relaxation of the dispersed phase of the <i>i</i> th bin (ms)
dmax	max, stable diameter (um)	Talm	log mean of transverse relaxation distribution (ms)
d_{y}	volume weighted mean diameter (um)	V_i	dimensionless viscosity group
d32	Sauter diameter (µm)	Ŵe	Weber number
E	energy input (k])	3	energy dissipation rate (J/s)
fi	amplitude of the <i>i</i> th bin (a.u.)	σ	interfacial tension (mN/m)
g	gravitational constant (m ² /s)	SR	surface relaxivity (µm/s)
Ĺ	impeller diameter (mm)	ρ_{CP}	continuous phase density (g/mL)
Ν	rotational speed of impeller (rpm)	ρ_{DP}	dispersed phase density (g/mL)
N_P	power number	μ_{CP}	continuous phase viscosity (Ns/m ²)
Р	power input (W)	μ_{DP}	dispersed phase viscosity (Ns/m ²)
Re	Reynolds number	φ	dispersed phase volume fraction
T_2	transverse relaxation (ms)	τ	time between $\pi/2$ and π pulses (µs)
$T_{2,bulk}$	bulk transverse relaxation (ms)		

drop breakup in turbulent flow that incorporates turbine mixers [7]. The Weber number, *We*, is defined according to the following equation:

$$We = \frac{\rho_{CP} N^2 L^3}{\sigma} \tag{3}$$

The density of the continuous phase is ρ_{CP} , the rotational speed of the impeller is *N*, the diameter of the impeller is *L*, and the interfacial tension is σ . Calabrese accounted for the viscosity of the dispersed phase and adjusted the correlation accordingly [8].

$$\frac{d_{32}}{L} = 0.053 W e^{-3/5} \left[1 + 0.97 V_i^{0.79} \right]^{3/5} \tag{4}$$

The viscosity group that accounts for the impact of the dispersed phase viscosity is given by V_i .

$$V_{i} = \left(\frac{\rho_{CP}}{\rho_{DP}}\right)^{1/2} \mu_{DP} \frac{NL}{\sigma}$$
(5)

The dispersed phase viscosity is represented by μ_{DP} .

To account for increased dispersed phase fractions, Calabrese developed an empirical formulation for dispersed phase fractions less than or equal to 0.2 [8].

$$\frac{d_{32}}{L} = 0.054(1+3\varphi)We^{-3/5} \left[1 + 4.42(1-2.5\varphi)V_i \left(\frac{d_{32}}{L}\right)^{1/3} \right]^{3/5}$$
(6)

The dispersed phase volume fraction is represented by φ . Eq. (6) accounts for the effect of dispersed phase volume fraction on disruptive energy.

A lack of data exists for concentrated emulsion formation in complex flows in the presence of surfactants, and there is presently a need for the direct measurement of drop size distributions of opaque emulsions such as crude oil emulsions. The NMR technique employed in this work fills that gap by directly measuring the emulsion droplet size distributions and transient behavior of concentrated crude oil emulsions. The direct measurements of real crude oil systems illustrate the complexities that arise in these systems.

2. Experimental methods

For all experiments presented in this paper, the dispersed phase was ASTM D1141 synthetic seawater (also referred to as brine in this paper) purchased from Ricca Chemical. The volume fraction was equal to 0.2 by combining 10 mL brine with 40 mL oil. Two

crude oils were used in this work, and the densities of all fluids used are contained in Table 1. The viscosities of the two oils were measured using a Brookfield rheometer for a range of temperatures as shown in Fig. 1. Fig. 2 shows the interfacial tensions for both crude oils in the presence of brine that were measured using the pendant drop technique. A KSV CAM 200 was used to perform the pendant drop measurements.

A six bladed turbine, also referred to as a Rushton turbine, was used to form the emulsions. This emulsification system was selected based on its ability to generate flow conditions in the turbulent regime [9]. A digital tachometer was used to measure the rotational speed of the impeller. The diameter of the impeller was 30 mm. The emulsions were prepared in a glass vessel with the inside diameter equal to 43 mm and the length equal to 210 mm. Fig. 3 shows the dimensions of the turbine relative to the glass vessel. The length of the rotor shaft was 198 mm, as shown in Fig. 4. The turbine blades were approximately 5 mm thick. For all experiments, the emulsions were mixed and measured in the same vessel. Therefore, the entire emulsion that was prepared was also used for the measurement. Because of the high viscosity of the crude oil B, the brine-in-crude-oil-B emulsions were formed in a circulating water bath with temperature equal to 303.2 K. A two sample Wheaton bench top roller was used to apply mild shear to select samples after emulsification. For all experiments for which mild shear was applied after emulsification, the rotational speed was set to 4 rpm. Table 2 summarizes the experimental conditions evaluated in this work. The Reynolds numbers reported in Table 2 were calculated according to the following equation [2,3].

$$Re = \frac{L^2 N \rho_{CP}}{\mu_{CP}} \tag{7}$$

Based on correlations by Rushton et al. [2,3], the power number, N_P , was determined and the power input to the system was calculated.

$$P = \frac{N_P}{\left(\frac{g}{N^2 L}\right)^{\left(\frac{a-\log(Re)}{b}\right)}} \frac{\rho_{CP} N^3 L^5}{g}$$
(8)

Table 1 Densities of the fluids.

- 2 - -

	Density (g/mL)
Synthetic seawater	1.03
Crude oil A	0.85
Crude oil B	0.91

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